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Project No.: 713

May 30, 2002

U.S. Nuclear Regulatory Commission
Attn: Document Control Desk
Washington, D.C. 20555

Subject: Submittal of Historical German Fuel Qualification Process Document

By this letter Exelon Generation Company (EGC) LLC submits the attached Pebble Bed Modular Reactor (PBMR) Pty. document, Number 000570-34, "Fuel Irradiation Qualification Report," which fulfills a commitment made by EGC during the March 28, 2002 pre-application meeting between the Nuclear Regulatory Commission and EGC. The document contains the historical German fuel qualification process referenced in EGC's PBMR fuel qualification test plan, which was provided to the NRC by letter dated March 18, 2002, "Document Supporting the March 28, 2002 Pre-application Meeting Regarding the Pebble Bed Modular Reactor (PBMR)." However, the information contained in the attachment does not supersede EGC's proposed PBMR fuel qualification test plan, nor does EGC request the NRC to perform an in-depth technical review of the material since EGC will be concluding pre-application activities.

If you have any questions or concerns regarding this matter, please contact R. M. Krich or me.

Sincerely,

Kevin F. Borton
Manager, Licensing

Attachment

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D064

Attachment

"Fuel Irradiation Qualification Report"
Document Number 000570-34

Submitted May 30, 2002

83 Pages

FUEL IRRADIATION QUALIFICATION REPORT

Document No.: 000570-34

Revision: 3

Programme Baseline: DDBL

US NRC VERSION

SUBMITTED ON MAY 30 2002

CONFIGURATION CONTROL

Item	Control Information
Programme:	PBMR
Project:	
Title:	Fuel Irradiation Qualification Report
Doc. Reference:	000570-34
Creation Date:	08/05/2002
Classification:	Confidential

DOCUMENT HISTORY

Rev.	Date	Author	Saved As	Changes
A?	07/01/2001	J H Venter		Original draft document
B	22/01/2001	J H Venter		Changes proposed during review
1	16/02/2001	J H Venter		Changes proposed during review and update of some test parameter values
2	03/04/2001	J H Venter		Changes and additions proposed after review by Eskom Client Office on 30/03/2001
3	09/05/2002	J H Venter	000570-34da2	General revision. Table 18 and Section 6 added; Figures 13 and 15 revised

DOCUMENT APPROVAL

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ABBREVIATIONS AND ACRONYMS

Abbreviation or Acronym	Definition
AVR	Arbeitsgemeinschaft Versuchsreaktor
BISO	Bi-coated Isotropic
BOL	Beginning of Life
DLOFC	Depressurized Loss of Forced Cooling
EDN	Equivalent DIDO Nickel
EOL	End of Life
FIMA	Fissions per Initial Metal Atoms
FPD	Full Power Days
HEU	Highly Enriched Uranium
HFR	High Flux Reactor (Petten)
HTI	High Temperature Isotropic
HTR	High Temperature Reactor
ILTI	Inner Low Temperature Isotropic
IVV-2M	Material Test Reactor at Zarechny, Russia
LEU	Low Enrichment Uranium
LTI	Low Temperature Isotropic
MWd/tU	Megawatt Day per Tonne Uranium
OLTI	Outer Low Temperature Isotropic
OTTO	Once Through Then Out
PBMR	Pebble Bed Modular Reactor
PIE	Post-irradiation Evaluation
PLOFC	Pressurized Loss of Forced Cooling
QA	Quality Assurance
R/B	Release-to-birth ratio
SiC	Silicon Carbide
TRISO	Tri-coated Isotropic
VLT	Voll Last Tagen (Equivalent Full Power Days)

1. SCOPE

This document describes the German fuel qualification process consisting of Phase 1 (Generic) and Phase 2 (Reactor Specific) irradiation qualification tests, and how the obtained data was used to define the design fission product release source for the HTR- Modul reactor. A comparison is made between PBMR and HTR-Modul operational conditions for fuel elements, and the applicability of German test results to PBMR fuel is evaluated. The results of the investigation are used to define a fuel irradiation test programme for PBMR.

2. INTRODUCTION

The starting point of the qualification strategy for PBMR fuel will be to use German HTR information as the basis for fuel manufacture. The same product and inspection specifications, critical hardware, and manufacturing processes that were used to produce German fuel will be used in the production of PBMR fuel, to ensure that it will be equivalent to German fuel. Thus PBMR fuel will be of proven design and manufactured according to known and accepted processes **[1]**.

The qualification programme for PBMR fuel can be split into two parts. The cold qualification will be carried out on as-manufactured and unirradiated fuel and will be done by the PBMR Fuel Division, while the fuel properties under actual reactor conditions will be verified through irradiation tests which will be done at a foreign laboratory under the supervision of PBMR personnel.

3. GERMAN FUEL DEVELOPMENT

3.1 Historical Review

Paragraphs 3.1 to **4.4.3** of this report are to all intents and purposes a free translation of **[2]**, with some explanation added where it was deemed necessary to promote better understanding.

The following characteristics of German pebble bed fuel have remained unchanged for all such fuel types produced in Germany since the first pebble bed spheres were irradiated in the Arbeitsgemeinschaft Versuchsreaktor (AVR) early in 1969. The design is characterized by:

- A graphite matrix, which is a mixture of natural graphite and electro -graphite (synthetic graphite) in the ratio 4:1 with a resinous binder.
- Spheres that are cold pressed under quasi-isostatic conditions.
- Spheres consisting of an inner fuel-containing region of 50 mm diameter with coated fuel particles distributed evenly in the graphite matrix.
- Fuel-containing region surrounded by a fuel-free region of thickness 5 mm consisting of the same matrix material as the fuel-containing region.
- Spheres heat-treated at a temperature of 1 950 °C, i.e. without any graphitization of the binder coke.

From about 1969, German pebble bed fuel design was aimed at producing fuel utilizing Highly Enriched Uranium (HEU) that could be used in the Uranium -Thorium fuel cycle. This cycle used a mixture of the carbides of highly enriched uranium and thorium.

In 1971, development was switched towards producing pebble bed fuel containing mixed oxides of uranium and thorium (U, Th) O_2. The invention of wet chemical procedures made it possible to develop spherical fuel kernels of this type for the first time. This development led to the design of the so-called HTI-BISO fuel particle (High Temperature Isotropic). In the BISO particle design, fuel kernels were coated with high-density pyrolitic carbon which was deposited in a fluidized bed by the dissociation of methane gas at a temperature of approximately 2 000 °C. BISO particles held the big advantage of a much-improved resistance to irradiation by high-energy neutrons due to the high degree of isotropy of the pyrolitic carbon coating.

In 1979/80, the uranium thorium fuel cycle was abandoned in favour of Low Enrichment Uranium (LEU) fuel without thorium. At the same time, the HTI-BISO particle design was replaced by the so-called LTI-TRISO (Low Temperature Isotropic) particle design. In this design, a layer of silicon carbide (SiC) was inserted between two pyrolitic carbon layers. SiC was deposited fro m methyltrichlorsilane (CH_3SiCl_3) at a temperature of 1 500 °C. As the SiC layer now performed the function of fission product retention, low- temperature pyrocarbon layers deposited from a C_2H_2/C_3H_6 mixture at 1 300 °C could replace the high-temperature pyrocarbon layers.

In 1982, an experimental programme for the generic qualification of UO_2-TRISO pebble bed fuel was started. This culminated in the reactor specific qualification of this fuel type for HTR- Modul that started in 1989.

3.2 Description of German Reference Fuel Element

The properties of TRISO coated particles are among the important factors determining the radiological safety of an operating pebble bed reactor. They determine the fission product retention in fuel elements, as well as the maximum fuel temperature that can be tolerated in the reactor core. For this reason, great care was taken to ensure that sufficient experimental evidence was accumulated during the qualification process, to support the design.

The components of the TRISO fuel element can be described in terms of the three main steps in the manufacturing process, i.e. kernel production, coated particle production, and fuel element production consisting of matrix preparation, pressing, and heat- treatment.

3.2.1 Kernel production

Nuclear fission takes place in a fuel kernel consisting of stoichiometric UO_2 of high density. In the fission process, a mixture of radioactive fission products is produced. Among these fission products are some gaseous as well as some volatile (mainly metallic) chemical elements. To ensure even stress loads on all fuel particles, a strict tolerance is placed on particle diameter (nominal value 500 μm). Furthermore, the production of highly spherical kernels will contribute to a reduction of stress peaks in the kernel coatings. Both requirements are met by using wet chemical procedures to form fuel kernels.

The starting material for kernel production is U_3O_8. The powder is dissolved in nitric acid and tetrahydro furfuryl alcohol is added. The solution is neutralized with ammonia and allowed to flow through an oscillating nozzle, which produces spherical droplets. The droplets fall through a gaseous ammonia atmosphere into an aqueous ammonia solution. While falling, the surface of a droplet gels and in the solution the droplet solidifies by conversion to ammonium di-uranate. Leaving them in the warm ammonia solution for some time ages the droplets. They are then washed, dried, calcined, reduced to UO_2 with hydrogen, and sintered to produce kernels.

3.2.2 Coated particle production

Four layers (coatings) surround each fuel kernel. The layers are deposited sequentially by dissociation of gaseous chemical compounds in a continuous process in a fluidized bed.

3.2.2.1 Buffer layer

The first layer in contact with the kernel is known as the buffer layer. It is deposited at a temperature of 1 000 °C from C_2H_2. The purpose of the buffer layer is to provide void volume for gaseous fission products in order to limit the pressure build-up within the coated particle. Conditions in the fluidized bed are arranged to keep the density of this layer below the maximum allowed value of 1.05 g/cm^3, which is approximately 46% of the theoretical density of pyrocarbon (2.26 g/cm^3). As a result of its porosity, the buffer layer also serves to decouple the fuel kernel, which swells with increasing burn-up of fuel, from the high-density outer layers, which are important in containing fission products within the coated particle.

3.2.2.2 Inner pyrocarbon layer

The inner high density, isotropic layer of pyrolitic carbon is also referred to as the ILTI (Inner Low Temperature Isotropic) pyrocarbon layer. It is deposited from a mixture of C_2H_2 and C_3H_6 at a temperature of 1 300 °C in the fluidized bed, and has a density of 1.9 g/cm ^{3}. This layer forms the first pressure barrier against the fission product pressure within the fuel kernel, thereby reducing the pressure on the next layer (SiC). Although intact LTI layers form a practically impenetrable barrier for fission gases and fission product iodine, they become increasingly pervious to caesium and strontium at temperatures above 1 200 °C.

3.2.2.3 Silicon carbide layer

When SiC is deposited from methyltrichlorsilane (CH_3SiCl_3) at 1 500 °C under the correct conditions, a layer of a minimum density 3.18 g/cm^3 (nearly theoretical density) is obtained, which is capable of practically complete retention of all fission products. The production of fuel elements having coated particles with intact SiC layers, and the guarantee that these layers will remain intact under all foreseeable reactor core conditions, form the basis for the safe operation of all pebble bed reactors.

Experiments have shown that SiC layers exposed to fission products, especially the platinum group metals (particularly Pd) and rare earth elements, for long times at temperatures above 1 600 °C, are chemically attacked by these elements. Fuel elements exposed to temperatures above 1 600 °C for times in the order of 100 h start releasing fission products such as caesium and strontium through intact SiC layers as a result of diffusion.

At temperatures in the region of 2 000 °C to 2 200 °C, thermal decomposition of SiC takes place and the SiC layer completely loses its fission product retention capability.

3.2.2.4 Outer pyrocarbon layer

The Outer Low Temperature Isotropic (OLTI) pyrolitic carbon layer is deposited in exactly the same way as the ILTI layer. The function of this layer is to protect the SiC layer against damage in the fuel manufacturing processes that follow. It also provides prestress on the outside of the SiC layer due to shrinkage under fast neutron irradiation during the fuel lifetime in the reactor core, thereby reducing the tensile stress in the SiC layer.

Experience has shown that coated particle failure in fuel elements could be greatly reduced by removing particles that show an unacceptable deviation from spherical shape. Unround coated particles show a much greater tendency to crack during isostatic pressing, than spherical particles.

3.2.3 Fuel element production

3.2.3.1 Fuel element matrix

Coated particles are embedded in matrix material consisting of a mixture of natural graphite and electrographite, together with a phenolic resin as binder material. Highly graphitized materials are used for fuel manufacture to ensure dimensional stability during irradiation with fast neutrons, because partially graphitized material undergoes further graphitization under fast neutron irradiation with accompanying dimensional changes. Highly graphitized material also has the desirable property that it can be pressed to the required density relatively easily.

Once a fuel element has been pressed, it is no longer possible to change the degree of graphitization of the materials contained in the fuel element. Graphitization takes place in the temperature range 2 700 °C to 3 000 °C, at which temperatures SiC is dissociated with the destruction of fission product retention capability. Even after the final heat- treatment at a temperature of 1 950 °C, each fuel element will contain some ungraphitized material originating from the resinous binder material, which is carbonized at 800 °C.

3.2.3.2 Overcoat

Before final pressing of a fuel element, a coating of matrix material is applied to each coated particle. This coating is known as the 'overcoat'. Its purpose is to prevent coated particles from coming into contact with each other, and thereby damaging their coatings during pressing of the fuel elements.

3.2.3.3 Pressing

Fuel elements are pressed at very high pressure without application of external heat, to obtain the required density that ensures adequate stability and heat conduction. This also provides the correct amount of carbon in the reactor core that determines heat capacity and moderation.

3.2.3.4 Fuel containing zone

Fuel particles are distributed evenly in the inner fuel-containing zone of diameter 50 mm, to prevent the development of hot spots in a fuel element.

3.2.3.5 Fuel-free zone

A fuel-free zone of thickness 5 mm, consisting of the same matrix material used for the inner zone, surrounds the inner fuel-containing zone of each fuel element. The purpose of this zone is to protect the inner zone from mechanical and chemical damage during handling and operation.

3.3 Fuel Element Properties

The most important specified properties for the reference HTR- Modul fuel element **[2]** are set out in **Table 1** to **Table 3** for fuel kernels, particle coatings and fuel elements. The 'Design Specification' column in the tables refers to Modul Design Values, while the 'Manufacturing Specification' column refers to values achieved during manufacture of EUO 2308 coated particles and test fuel elements. These particles and test fuel elements containing these particles were used for experiments HFR-K3 and FRJ2-K13 in Phase 1 of the irradiation qualification programme for HTR-Modul.

Experiments HFR-K3 and FRJ2-K13 provided the database from which fuel element behaviour under normal and upset event reactor conditions could be deduced. EUO 2308 particles were also used in the other Phase 1 irradiation experiments, as well as in charge AVR-19 loaded into the AVR reactor in July 1982.

From a comparison of the two specification columns, it is clear that the design specification was met for each specified property, including the very important 'defective SiC fraction'. This fraction contains all uranium not covered by an intact SiC layer. The fraction includes uranium contamination of the graphite matrix and particles with cracked or broken SiC layers, although their pyrocarbon layers may still be intact, as determined by the burn-leach method. Experience has shown that the minor deviations, because of technical requirements, for U235 enrichment and heavy metal content, do not invalidate experimental results in any way.

The design specifications also contain requirements relating to chemical and mechanical interactions between a fuel element and its surroundings during reactor operation. Test procedures, based on reactor operational conditions, are provided in quality control procedures to prove compliance of fuel elements with these specifications. Thus mechanical strength of fuel elements is specified in terms of breaking strength between parallel steel plates as well as the number of falls through a height of 4 m onto a pebble bed without breaking.

Dust generation in the primary circuit due to abrasion of fuel elements as they rub against each other is specified in terms of mass loss experienced when 20 fuel elements are rotated in a drum for 100 h. Corrosion resistance of fuel elements is specified in terms of mass loss experienced when fuel elements are exposed for 10 h to streaming helium gas containing one volume percent of water vapour at a temperature of 1 000 °C.

Experience has shown that fuel element properties such as mechanical strength, abrasion and corrosion resistance are not influenced to any great extent by irradiation **[9]**.

In **Table 1** to **Table 3** the symbols used have the following meanings:

- x = Single Value
- [x] = Average Value
- s = Standard Deviation

Table 1: Specifications for UO_2 Kernel

Parameter	Unit	Design Specification [2]	Manufacturing Results EUO 2308
U235 Enrichment	%	7.8	1)
Diameter	μm	[x] - s ≥ 480 [x] + s ≤ 520	[x] = 497 s = 14.1
Density	g/cm^3	x ≥ 10.4	[x] = 10.81

1) Variations in this parameter value were necessary to suit specific applications. These deviations have no influence on test results.

Table 2: Specifications for Coated Particles

Parameter	Unit	Design Specification [2]	Manufacturing Results EUO 2308
Buffer Layer:			
Thickness	μm	[x] – s ≥ 72 [x] + s ≤ 108	[x] = 94 s = 10.3
Density	g/cm^3	[x] ≤ 1.05	[x] = 1.00
Inner High Density Pyrocarbon Layer:			
Thickness	μm	[x] – s ≥ 30 [x] +s ≤ 50	[x] = 41 s = 4.0
Density	g/cm^3	[x] = 1.90 ± 0,1	Not Measured
SiC Layer:			
Thickness	μm	[x] – s ≥ 31 [x] + s ≤ 39	[x] = 36 s = 1.7
Density	g/cm^3	[x] ≥ 3.18	[x] = 3.20
Outer High Density Pyrocarbon Layer:			
Thickness	μm	[x] – s ≥ 30 [x] +s ≤ 50	[x] = 40 s = 2.2
Density	g/cm^3	[x] = 1.90 ± 0.1	[x] = 1.88

Table 3: Specifications for Fuel Elements/Matrix

Parameter	Unit	Design Specification [3]	Manufacturing Results EUO 2308
Thickness of Fuel-free Zone	mm	$x = 5 \pm 1$	$x > 5$
Matrix Density	g/cm^3	$[x] = 1.75 \pm 0.02$	$[x] = 1.75$
Thermal Conductivity at 1 000 °C	W/(m.K)	$[x] \geq 25$	$[x] = 35$
Corrosion Rate at 1 000 °C [2)]	$mg/(cm^2.h)$	$[x] \leq 1.3$ $x \leq 1.5$	$[x] = 0.99$ $x \leq 1.07$
Thermal Expansion Anisotropy	Ratio	$[x] \leq 1.3$	$[x] = 1.13$
Breaking Strength[3)]	kN	$x \geq 18$	$x \geq 20.6$
Fall Resistance[4)]	Number of Falls	$x \geq 50$	$x \geq 62$
Heavy Metal Content	g/FE	$[x] = 7.0$	1)
U235 Content	g/FE	$[x] = 0.546$	1)
Defective SiC[5)]	Ratio	$[x] \leq 6 \times 10^{-5}$	$[x] = 3.5 \times 10^{-5}$

1) Variations in these parameters were necessary to suit specific applications. These deviations have no influence on test results.

2) Standard test conditions: 10 h in streaming helium at 1 bar containing 1 vol% of water.

3) Pressed between parallel steel plates.

4) Number of falls through a height of 4 m onto a pebble bed without breaking.

5) Includes all uranium in a fuel element that is not enclosed within an intact SiC layer, as determined by the burn-leach test method.

4. GERMAN FUEL IRRADIATION QUALIFICATION PROGRAMME: PHASE 1

4.1 Irradiation Qualification Concept

In Germany, a number of pebble bed reactor concepts were developed around the LEU-TRISO fuel element design. Therefore irradiation qualification of these fuel elements was done in two phases.

- Phase 1 was a generic phase during which prototype material was irradiated under conditions that covered the whole range of expected parameters envisaged for all reactor concepts based on LEU-TRISO fuel. Thus Phase 1 experiments provided the basic data needed to determine fission product release source terms under all foreseeable reactor conditions.
- Phase 2 experiments were reactor specific (HTR- Modul), using typical production material and simulating actual reactor operation conditions as closely as allowed by the materials testing reactor that was used for irradiation. Thus Phase 2 experiments were designed to demonstrate the transferability of Phase 1 data to the HTR-Modul concept, and also to widen the statistical database on fission product release for HTR- Modul.

4.2 Phase 1 Experiments

The main purpose of irradiation measurements was to identify and to quantify all mechanisms that could possibly contribute to the release of fission products from fuel elements. As a result of the negligible uranium contamination of TRISO coated particles manufactured by NUKEM and the excellent containment properties of intact coated particles for most fission products, the most important fission product release source is the release from defective coated particles. The most important factors contributing to failure of coated particles are:

- production processes;
- irradiation by fast neutrons; and
- temperature.

The failed particle fraction resulting from each of these factors can be measured through quality control measurements, irradiation data, and post-irradiation heating experiments respectively.

If uranium contamination of coated particles and matrix graphite is negligible, the only remaining sources of fission product release are diffusion from intact coated particles and from defective coated particles. Irradiation experiments were designed to investigate both of these release mechanisms with varying temperature.

The experiments performed for LEU-TRISO reference coated particles are shown in **Table 4** . All tests shown in **Table 4** used coated particles from lot EUO 2308 (NUKEM/HOBEG).

Table 4: Purpose of Phase I Irradiation Experiments for LEU-TRISO Fuel

Purpose	Irradiation Experiment LEU-TRISO Phase I
Coated Particle Quality (Failed Particle Fraction)	HFR-P4 SL-P1
Fuel Element Quality	HFR-K3
Fission Product Transport from: - Intact Particles - Defective Particles - Fuel Elements (AVR small-scale test)	 FRJ-P27 FRJ2-P28 FRJ2-K13
AVR large-scale test	AVR-19 AVR-21

For technical reasons, not all irradiation experiments were conducted with full-size fuel elements. In tests HFR-P4 and SL-P1, so-called 'small spheres' were used. These spheres were produced using identical materials and processes to those used for normal fuel elements. However, the spheres had a fuel containing zone of only 20 mm diameter and a fuel free zone of thickness 20 mm. Cylindrical test samples that would fit into the test rigs were machined from these small spheres. The irradiation rigs used in irradiation experiments in Jülich (FRJ2) were designed for irradiating compacts. Cylindrical compacts of diameter 28 mm and height 30 mm with a higher coated particle density than fuel elements were hot-pressed in matrix graphite in a steel die. As a result of these deviations in geometry, production method and materials from fuel elements, the bedding of particles in the test samples was not strictly representative of that in fuel elements. Nevertheless the effect of these deviations was considered to be minimal, and was neglected.

After temperature, the most important irradiation target for Phase 1 experiments was burn-up. The reason was that coated particle failure during reactor operation is determined by tensile stress in the SiC layer, which is caused by burn-up dependent fission product pressure within the coated particles. It was also known from previous irradiation experiments on HTI-BISO coated particles that fast neutron fluence, in combination with temperature effects, caused failure of high-density pyrolitic carbon coatings.

It is unavoidable that a certain number of anisotropic pyrolitic carbon coatings are produced during the coating process. Irradiation by fast neutrons causes further anisotropic structural changes in these particles, which induces tensile stress and causes failure.

For Phase 1 experiments the target burn-up was set at 10% to 12% Fissions per Initial Metal Atoms (FIMA) which corresponds to 90 000 to 107 500 Megawatt Day per Tonne Uranium (MWd/tU). The average and maximum burn-ups for HTR-Modul were 8.93 and 9.83%FIMA, which corresponded to 80 000 and 88 000 MWd/tU respectively.

A fast neutron fluence of 6 $x10^{21}$ n/cm^2 (E > 0.1 MeV) was specified for those experiments designed to measure failed particle fractions, i.e. HFR-P4, SL-P1 and HFR-K3. For HTR- Modul, the average and maximum fast neutron fluence were 2.1 x 10^{21} and 2.4 x 10^{21} n/cm^2 respectively.

To investigate the temperature dependence of particle failure, irradiation temperatures of 1 000 °C and 1 200 °C were specified for most of the tests, although values of 800 °C and 1 300 °C were specified for a few tests. The irradiation temperature was kept constant for the whole duration of the test in most cases. An exception was test FRJ-P28 that contained a number of

artificially introduced defective particles. The temperature for this test was varied in order to determine the temperature dependence of fission gas release in more detail.

In HTR-Modul, a fuel element would circulate through the core 15 times and the maximum temperature reached in any one cycle was 865 °C. Power variations, similar to temperature variations, are experienced by fuel elements as they are cycled through the reactor core. During irradiation experiments, a nearly constant power of 4 kW per sphere was generated, which corresponded to a power of 250 mW per coated particle. In HTR-Modul, the maximum power during a single cycle was 1.40 kW per sphere, which corresponded to 125 mW per coated particle.

Taken together, the target values for LEU-TRISO Phase 1 experiments enveloped and, in some cases, exceeded the design limits for HTR-Modul.

4.3 Performance of Phase 1 Measurement Programme

The performance of Phase 1 experiments will be discussed under the following headings:

- Design of experiments.
- Experimental samples used.
- Irradiation.
- Post-irradiation Evaluation (PIE).

Details of Phase 1 experiments are provided in the form of tables, with descriptions of the different parameters in the tables in the paragraphs following the tables.

4.3.1 Design of Phase 1 experiments

The Phase 1 irradiation programme consisted of six experiments performed in three different reactors. The design details are shown in **Table 5**. Experiments were allocated to different reactors on the basis of the technical capabilities available at each reactor.

Experiments requiring full burn-up and fast neutron fluence were performed in the HFR Petten (HFR-P4 and HFR-K3) and in SILOE Grenoble (SL-P1).

Experiments requiring priority investigations regarding fission product release from intact particles (FRJ2-P27) and fuel elements (FRJ-K13) as well as defective particles (FRJ2-P28) were performed in FRJ2 in Jülich. For these irradiation experiments it was not essential to reach the full target value for fast neutron fluence.

Irradiation experiments were carried out at temperatures of 800 °C, 1000 °C and 1 200 °C. Temperatures were kept constant for the full duration of the tests. As such the experiments were more representative of once Through Then Out (OTTO) cycle conditions than of multiple cycle conditions that would be encountered in HTR-Modul. In experiment FRJ2-P28, the temperature was varied to study fission product release as a function of temperature.

Coated particles of lot EUO 2308 manufactured by NUKEM/HOBEG were used in all Phase 1 experiments. The defective particles introduced artificially in experiment FRJ2-P28 consisted of similar fuel kernels to those used to produce EUO 2308 coated particles taken from kernel charge UOS 331. The defective particles were coated with only a buffer layer, and then removed

from the coater. By providing kernels with a buffer layer, it was ensured that fission products released from the kernel by recoil were slowed down and transpo rted in exactly the same way as fission products produced in kernels whose high density coatings have become defective.

Table 5: Phase 1 LEU-TRISO Irradiation Programme – Experiments and Purpose

Experiment Number	Capsule Number	Particle Lot	Samples: Number, Shape	Nominal Target Values			General Purpose
				Fuel Temperature °C	Burn-up %FIMA	Fast Neutron Fluence (> 0,1 MeV) x 10^{21} n/cm^2	
HFR-P4	A/01 C/03	EUO 2308 EUO 2308	12 small spheres 12 small spheres	1 000 1 200	Max. 12 Max. 12	Max. 6.2 Max. 6.2	Failed particle fraction as function of burn-up, fast neutron fluence and temperature for medium/ upper temperature range
SL-P1	-	EUO 2308	12 small spheres	800	Max. 12	Max. 6	As for HFR-P4 for lower temperature range
HFR-K3	1/A (bottom) 2/B (middle) 3/C (top)	EUO 2308 EUO 2308 EUO 2308	1 fuel element 2 fuel elements 1 fuel element	1 200 1 000 1 200	Approx. 8 10 Approx. 8	6.2 6.2 6.2	Reference test with full fuel element at maximum burn-up and maximum fast neutron fluence
FRJ2-K3	1 (top) 2 (bottom)	EUO 2308 EUO 2308	2 fuel elements 2 fuel elements	Approx. 1 200 Approx. 1 200	10 10	- -	Parallel test to AVR partial charge XIX for investigation of fission product transport
FRJ2-P27	1 (top) 2 (middle) 3 (bottom)	EUO 2308 EUO 2308 EUO 2308	3 compacts, 2 coupons 3 compacts, 2 coupons 3 compacts, 2 coupons	900 1 300 1 100	8 10 8	Approx. 2 Approx. 2 Approx. 2	Fission product transport test at different temperatures for full target burn-up
FRJ2-P28	1 (top) 2 (middle) 3 (bottom)	EUO 2308 EUO 2308 EUO 2308	3 compacts, 2 coupons 3 compacts, 2 coupons 3 compacts, 2 coupons	800 1 200 1 000	10 12 10	Approx. 2 Approx. 2 Approx. 2	Fission product release from defective particles at different temperatures (1% of particles without high density layers)

Explanation of Table 5 Column Headings:

a. Experiment Number

Experiment number consists of the following:

Abbreviation for the test reactor	HFR for HFR at Petten
	SL for SILOE at Grenoble
	FRJ2 for FRJ2 'DIDO' at Jülich
Test sample type	P for samples containing particles, i.e. deviating in form and bedding from fuel elements
	K for full-size fuel elements

Number for the specific test at the specific test reactor.

b. Capsule Number

The capsule number provided the orientation of the test sample relative to the irradiation rig used. A short description of each rig is provided below:

- HFR-P4: 'TRIO' rig with three thin parallel capsules over the active height of the rig numbered A/01, B/02, and C/03. Each capsule took 12 small spheres numbered 1.1 to 1.12, 2.1 to 2.12 and 3.1 to 3.12 from top to bottom.
- Capsule B/02 of test HFR-P4 contained HOBEG particles of lot EUO 2309 with nominally 50 μm SiC layers, which were not considered in the analysis following.
- SL-P1: 'TUMULT' rig consisting of a thin capsule that could accommodate small spheres or cylindrical probes.
- HFR-K13: 'BEST' rig for fuel element irradiation in HFR. For these experiments, it had three capsules. The top and bottom capsules contained one fuel element each, while the middle capsule contained two fuel elements. The middle elements were placed in the maximum neutron flux position, while the outer elements were in positions of lower flux.
- FRJ-K13: The rig used was the Jülich Sphere rig consisting of two separate capsules containing two full-size fuel elements each.
- FRJ2-P27/28: The rig consisted of three separate capsules containing three compacts and two coupons per capsule.

In all irradiation experiments, the individual capsules were swept with carrier gas to measure released fission gases. The temperatures of individual capsules could be controlled separately.

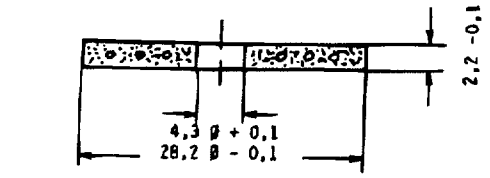
c. Particle Type

NUKEM/HOBEG particle lot EUO 2308 coated particles were used in all capsules in the irradiation tests. The properties of this coated particle type are shown in **Table 6** . The only exception was capsule B/02 in the HFR-P4 test, which contained particle type EUO 2309 coated particles. These particles had 50 μm thick SiC layers. The purpose of the thicker SiC layer was to investigate the improvement in the retention of fission products by increasing the thickness of the SiC layer. Capsules 1 and 3 of FRJ-P27 also contained one coupon each (34 coated particles per coupon) containing type EUO 2309 coated particles.

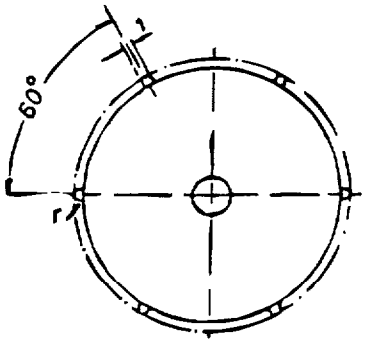
d. Samples: Number and Shape

For technical and financial reasons, full-size fuel elements could not be inserted into all irradiation rigs. The sample geometry and the number of samples inserted into each capsule were determined by the geometry of the irradiation position available in each reactor that was utilized.

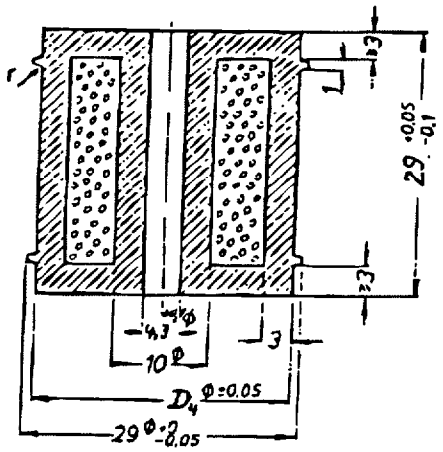
- Fuel Element: A full-sized fuel element sphere of outer diameter 60 mm with full coated particle and heavy metal loading.
- Small Sphere: Small spheres were manufactured using the same methods and materials used to manufacture fuel elements. However, they had a fuel zone of 20 mm diameter with a fuel-free layer of thickness 20 mm. Cylindrical samples were machined from small spheres. For tests HFR-P4 and SL-P1, cylinders of 24 mm to 30 mm diameter, depending on heat transfer design requirements, and 32 mm height were loaded into the irradiation capsules.



Coupon



Compact



Capsule	1 900°C			2 1300°C			3 1100°C		
Compact-Nr.	2	3	4	9	10	11	14	15	17
D_4-Ø mm	27,6	28,2	28,3	27,0	27,6	27,4	28,2	28,2	28,0

Figure 1: Irradiation Sample Types for Experiment FRJ2-P27

- Compact: Annular cylinder with fuel-free region surrounding the fuel-containing region as shown in **Figure 1**. The cylinder was hot pressed. As a result of the different methods and materials used in manufacture, the bedding of coated particles in compacts was not strictly representative of normal fuel particles. The effect of this difference was considered to be negligible. The compacts of test FRJ2-P28 were loaded with coated particles, 1% of which were defective.
- Coupon: A coupon was a thin pressed disk containing a number of coated particles arranged in a circle as shown in **Figure 1**. This design was ideally suited to the investigation of individual coated particles before and after irradiation. In test FRJ2 -P27, two coupons containing 34 particles each of type EUO 2309 were inserted between the three compacts. In test FRJ2-P28, five defective particles of lot EUO 2308 each were pressed into all six coupons. These defective particles had kernels and buffer layers identical to lot EUO 2308 coated particles, but were not coated with any high-density layers
- .

e. Nominal Target Value

The LEU-TRISO programme was designed to cover all expected irradiation requirements for all HTR reactor types under consideration at the time. The fast neutron fluence requirement for HTR-Modul was quite modest in comparison with some other reactor types, and thus its requirements were adequately covered in the experimental programme.

- Fuel Temperature: Capsule temperatures for single capsules were kept constant within the smallest possible ranges during irradiation. For FRJ2-P28, an additional temperature programme was followed at the start and end of the irradiation to study the temperature and burn-up dependence of fission gas release from defective particles.
- Burn-up: The values stated are for end of irradiation heavy metal burn -up. For HFR-P4 and SL-P1, these were for the probes with the highest burn-up within the capsule arrangement. The irradiation target was the state d burn-up value, or otherwise a particle failure fraction of approximately 0.5% as measured using fission gas release.
- Fast Neutron Fluence (E > 0,1 MeV): The target fast neutron fluence of at least 6×10^{21} n/cm^2 was not valid for irradiation in FRJ2, because of the relatively low fast neutron flux available in the reactor, especially in the core boundary position that was used for FRJ2-K13. For purposes of investigating fission product transport, which is dominated by burn-up and temperature, the effect of fast neutron fluence was negligible. Furthermore, particle failure in TRISO particles is caused primarily by burn-up, which is related to the thermal neutron fluence. It is burn-up and the resultant fission gas pressure that eventually cause particle failure due to excessive tensile stress generated in the SiC layers of coated particles.

f. General Purpose

The main purpose of each experiment is described in this column using only keywords and phrases. Each experiment though, provided a whole range of results that were not specified in the General Purpose column. For example, all experiments yielded values for free uranium fraction of as-manufactured fuel or the equivalent quantity 'fission product release at start of irradiation'.

Experiment FRJ2-P28 was not considered in the statistical analysis, because it contained deliberately introduced defective particles, which was not representative of LEU-TRISO coated particles.

Table 6: Phase 1 LEU-TRISO Irradiation Programme – Coated Particle Lot EUO 2308 (Measured Values)

HFR-K3/FRJ2-K13					
1. Uncoated Particles: UOS 331					
Enrichment U-235		%	9.82		
U Content	O/U Ratio	%	%U: 88.4	O/U: 2.006	
Sieve Fraction *		µm	450 – 560		
Density		$g.cm^{-3}$	10.81 (Hg density measurement)		
Diameter:		µm	Measured on approx. 3 200 particles		
Average			497		
Standard Deviation			14.1		
95/95 Boundary values			469/525		
Particle Shape:		Piece			
dM/dm ≤ 1,2			1 measured in 1 000 particles		
dM/dm ≤ 1,5			0 measured in 1 000 particles		
Other Forms			0 measured in 2 000 particles		
Impurities					
		ppm	Ag: < 0.05	Gd: < 0.02	
			B: < 0.08	Li: < 1	
			Ca: 640	Mg: 0.6	
			Cd: < 0.07	Mn: < 1	
			Cl: < 3	Mo: < 1	
			Co: < 1	Ni: 3	
			Cr: < 3	Sm: < 0.04	
			Cu: < 0.3	Sn: < 5	
			Dy: < 0.02	Ti: < 0.2	
			Eu: < 0.02	V: < 0.2	
			Fe: 17	Zn: < 20	
2. Coated Particles: EUO 2308					
Coated Particle Mass per Batch*		g	5 000		
			Layer 1	Layer 2	Layer 3	Layer 4
Coating Gas*			C_2H_2	C_2H_2/C_3H_6	CH_3SiCl_3	C_2H_2/C_3H_6
Layer Thickness:						
Average		µm	94 **	41 **	36 ***	40 ***
Standard Deviation			10.3	4.0	1.7	2.2
Measured on		Particles	100	100	100	100
Layer Density		g/cm^3	1.00		3.20	1.88
Anisotropy BAF from OAF				1.053		1.019

HFR-K3/FRJ2-K13		
Sieve Fraction*	μm	800 – 1 000
Geometric Density	g/cm^3	3.48
U Content	mass%	47.72
U Surface Contamination	U/U_{total}	0.06×10^{-6}
Defective SiC Layers	U/U_{total}	$< 0.5 \times 10^{-7}$
Average Particle Diameter	μm	895 s = 28.9

Note: In **Table 6**, the asterisks denote the following:

* Production Specification.

** Metallography.

*** Microradiography.

4.3.2 Experimental samples used

The loading of capsules and the identification of irradiation samples are shown in **Table 7**. Experiments HFR-P4 and SL-P1 used identical 'small spheres' as described in ***paragraph 4.3.1***. Experiments FRJ2-P27 and FRJ-P28 used cylindrical compacts and coupons while full-sized fuel elements were used in experiments HFR-K3 and FRJ2-K13.

During quality control measurements on fuel used for the test samples, the failed particle fraction was one of the properties measured using the 'burn-leach' method. This method measure all uranium not covered by an intact SiC layer, and it includes failed particles as well as uranium contamination of the outer pyrolitic carbon layer and of the graphite matrix. Since this is a destructive method, it was performed on separate samples taken from the same batch as the samples used for irradiation tests.

Table 7: Phase 1 LEU-TRISO Programme – Capsule Loading and Characterization

Experiment Number	Capsule	Sample Number and Form	Sample Loading		Number of Particles in		Measurement 'Defective SiC'				
			Uranium (gram)	Particles	Capsule	Test	Single Values U_{free}/U_{total}	Number of Measure-ments	Particles Investigated	U/Particle U/Sample	Number of Defective Particles
HFR-P4	A/01	12 small spheres	1.018	1 631	19 572		$< 1 \times 10^{-6}$				0
	C/03	12 small spheres	1.018	1 631	19 572	39 144	$< 1 \times 10^{-6}$				0
							$<1 \times 10^{-6}$	5	8 155	6.09×10^{-4}	0
SL-P1	-	12 small spheres	1.018	1 631	19 572	19 572	$<1 \times 10^{-6}$				0
							$<1 \times 10^{-6}$				0
HFR-K3	1/A	1 fuel element	10.22	16 400	16 400		59×10^{-6}				1
	2/B	2 fuel elements	10.22	16 400	32 800		$< 1 \times 10^{-6}$				0
	3/C	1 fuel element	10.22	16 400	16 400	65 600	113×10^{-6}	5	82 000	6.11×10^{-5}	2
FRJ2-K13	1	2 fuel elements	10.22	16 400	32 800		$< 1 \times 10^{-6}$				0
	2	2 fuel elements	10.22	16 400	32 800	65 600	$< 1 \times 10^{-6}$				0
FRJ-P27	1	3 compacts, 2 coupons	1.515	2 424	7 340		$< 3 \times 10^{-6}$				0
			0.021	34							
	2	3 compacts, 2 coupons	1.515	2 424	7 340		$< 3 \times 10^{-6}$				0
			0.021	34							
	3	3 compacts, 2 coupons	1.515	2 424	7 340	22 020	$< 3 \times 10^{-6}$	5	12 120	4.09×10^{-4}	0
			0.021	34							
							$< 3 \times 10^{-6}$				0
							$< 3 \times 10^{-6}$				0

Explanation of Table 7 Column Headings:

The headings of the first three columns have exactly the same meaning as in **Table 5** .

a. Sample Loading

The sample loading was specified in terms of the heavy metal content and number of particles in each test sample, which was taken from the applicable pre -irradiation report. For FRJ2-P27, a further distinction was made between the loading for compacts and coupons. It should also be remembered that one coupon in capsules 1 and 3 of FRJ2 -P27 contained particles of lot EUO 2309.

b. Number of Particles in Capsule/Test

The total number of particles per capsule was calculated from the particle loading per sample and the number of samples per capsule. In the case of test FRJ2 -P27, particles of lot EUO 2309 were included in this number.

In total, excluding FRJ2-P28 and AVR fuel elements, 212 000 particles of lot EUO 2308 were irradiated. It should be remembered that the calculations involving these large numbers of coated particles are really dealing with the heavy metal content of single coated particles and of the samples containing the particles.

In the pre-irradiation report for SL-P1, the number of coated particles per small sphere was stated as 1 666. However, these probes came from the same production lot as those used in HFR-P4 which contained 1 631 particles per small sphere, and this value was used for SL-P1.

c. Measurement 'Defective SiC'

During quality control measurements on fuel used for the test samples, the failed particle fraction was one of the properties measured using the 'burn-leach' method. This method measures all uranium not covered by an intact SiC layer, and it includes failed particles, uranium contamination of the outer pyrolitic carbon layer and of the graphite matrix. Since this is a destructive method, it was performed on separate samples taken from the same batch as the samples used for irradiation tests. The test samples for HFR- P4 and SL-P1 were taken from the same batch, and similarly those used in HFR-K3 and FRJ2-K13. Compacts and coupons were manufactured especially for FRJ2 -P27. Therefore characterization of these three sample types was done collectively. For instance, the five measurements for small spheres shown **Table 6** were valid for both HFR-P4 and SL-P1, and similarly for the fuel elements used in HFR -K3 and FRJ2-K13.

The five measured values determined for HFR-P4/SL-P1 parallel samples and FRJ2-P27 yielded results that were below the detection limit. The detection limits for the samples were different because of the different coa ted particle loading of the probes. By comparing the measured value with the value that would result from a single defective particle in a sample, the number of defective particles could be determined from the ratio U/Particle : U/Sample. From the results for the three tests, it could be stated that not one of the samples contained a defective particle. In two of the parallel samples for HFR-K3/FRJ2-K13, values exceeding the detection limit were found. These values corresponded to one and two defective particles respectively. Thus in the 15 samples investigated (102 000 coated particles), three defective particles were found, which corresponds to a free uranium fraction of 3×10^{-5} for as-manufactured fuel. To put this value into perspective, it must be stated that all the samples for these tests were manufactured before the sorting of unround particles after

application of the outer pyrolitic coating was introduced into the manufacturing process. The sorting process greatly reduced the number of defective particles resulting from pressing.

In assessing the 'free uranium' fraction, also known as 'defective SiC' fraction, all uranium that can be measured using the quality control method known as 'burn-leach test' is included. In this method the sample, be it loose coated particles, small spheres, compacts or fuel elements, is burned at approximately 800 °C and then treated with nitric acid. The uranium content of the acid is measured. Using this method, all uranium that is not protected by an intact SiC layer is measured. This includes:

- defective particles with damaged high-density LTI and SiC coatings;
- particles with defective SiC coatings although the PyC layer might be intact; and
- uranium contamination of the outer PyC coating and fuel element matrix.

As can be seen from **Table 7**, the 'defective SiC' ratios for the small spheres used in experiments HFR-P4 and SL-P1, as well as for the compacts used in experiment FRJ2-P27, were below the single defective particle value of the burn -leach test at 1×10^{-6} to 3×10^{-6}. This meant that in those samples, not a single defective particle was detected. The method would have given a value of 4.1×10^{-4} to 6.1×10^{-4} for the samples used, if a single defective particle was present (refer to column $\frac{U\,/\,particle}{U\,/\,sample}$ in **Table 7**).

For three of the five parallel samples for the HFR- K3 and FRJ2-K13 experiments, the defective SiC ratio was below the single defective particle value of the burn-leach test. The measured values for the other two samples in these tests (59×10^{-6} and 113×10^{-6}) indicated that one fuel element had a single defective particle, and the other fuel element had two defective particles out of nominally 16 400 particles per fuel element.

These results confirmed previous experience that measured free uranium for fuel elements containing TRISO coated particles could be attributed entirely to single defective particles, while uranium contamination contributed an almost negligible amount.

Contamination of the outer pyrolitic carbon layer, which could lead to fission product release, was practically non-existent. It can be seen from **Table 6** that the free uranium fraction measured by burn-leach for loose particles of lot EUO 2308 was determined as less than 5×10^{-8}.

The uranium contamination of matrix graphite was also very low and could be traced to the natural uranium (and thorium) remaining after the graphite has been refined and pu rified. It was known from previous investigations that the natural uranium concentration in matrix graphite ranged between 50 ng to 200 ng per gram carbon, which corresponds to 10 μg to 40 μg of natural uranium per fuel element. Taking into account the uranium inventory of the HFR-K3/FRJ2-K13 test fuel elements of 10.22 g U per element, the free uranium fraction due to matrix contamination was 1 to 4×10^{-6}, which was also of the order of the detection limit for burn - leach. These facts proved that practically no additional contamination of the matrix took place during any manufacturing step in the production of fuel elements.

Taken together, the measured defective SiC values using burn-leach, predicted a defective SiC fraction of 2.85×10^{-5} for fuel elements using LEU-TRISO particles of lot EUO 2308 under normal operational conditions. This value was very close to the value of 2.93×10^{-5} which was calculated using the fact that three failed particles were detected from a total of 102 275 investigated. Thus the free uranium fraction for fresh fuel was taken to be 3×10^{-5}.

Furthermore, all Phase 1 experiments were performed before the removal of unround particles by means of a vibration plate was incorporated in the manufacturing process. Removal of these particles was expected to result in a further reduction of the number of particles becoming defective during the pressing process.

4.3.3 Irradiation

The irradiation details for Phase 1 experiments are shown in **Table 8** . In comparing the target values of **Table 4** with the actual values reached in **Table 8**, it was clear that there were some discrepancies. The actual values could only be measured after completion of the irradiation by means of gamma and mass spectrometry. During irradiation, calculated values were used to determine when experiments should be terminated on reaching target values. However, some burn-up values were higher and some were lower than the target values. These deviations were related to the neutron spectrum at the irradiation positions used. The irradiation of experiment HFR-P4 was done in a core position with a relatively hard neutron spectrum, and showed a markedly higher burn-up than the calculated value indicated.

The measured and calculated values for test HFR-K3 irradiated at the core boundary corresponded well. The measured burn-up values for tests FRJ-P27 and FRJ-P28 performed in a DIDO core position with a relatively soft spectrum were well below the calculated values. These negative deviations were even more marked for test FRJ2-K13, where the irradiation was performed in a reflector position in DIDO.

In most of Phase 1 irradiation experiments, fission gas release rates were very low. This was true for all capsules of HFR-P4, HFR-K3, FRJ2-K13, and for capsule 3 of experiment FRJ2-P27. The Release-to-birth (R/B) ratio for the fission gas Kr88 was in the order of 10^{-9} to 10^{-8} at start of irradiation (Beginning of Life [BOL]). For these capsules, the R/B value increases slowly with time, reaching a maximum value of 3×10^{-7}. This slow increase in R/B could be attributed to breeding effects in natural uranium and thorium present in matrix graphite.

The higher initial fission gas release rate for test SL-P1 and capsules 1 and 2 of test FRJ-P27 could be attributed to defective particles present in the fresh fuel. The number of defective particles in these fuel elements could be determined using the results obtained for the release of fission gas from defective particles, which were measured as a function of temperature in experiment FRJ2-P28. Using the experiment FRJ2-P28 determined released rates for particles and the total number of particles present in each capsule, an R/B value was calculated for a single defective particle. The number of defective particles could then be calculated by dividing the measured R/B value by the R/B value for a single defective particle. Using this method, the number of defective particles was estimated as five for SL-P1, and five and three respectively in capsules 1 and 2 of test FRJ-P27. This estimate was considered to be conservative due to the relatively large variations in temperature measurements. On similar grounds, the presence of defective particles in the capsules with low fission product release rates could be excluded. Refer to **Table 9** for results of Phase 1 irradiation tests.

Table 8: Phase 1 LEU-TRISO Programme – Irradiation Data

Irradiation Experiment	Capsule Number	Irradiation			Burn-up	Fast Fluence (E>0,1 MeV) n/cm^2 (x 10^{21})	BOL				EOL			
							Temperature °C		R/B		Temperature °C		R/B	
		Start	End	Dura-tion FPD			Surface	Fuel Element	Kr85m	Kr88	Surface	Fuel Element	Kr85m	Kr88
HFR-P4	A/01 C/03	10.06.82	22.11.83	350.7	10.9 to 14.7	6.0 to 8.0	960 to 1 030		$3.5.10^{-9}$ $3.6.10^{-9}$	$3.6.10^{-9}$ $2.5.10^{-9}$	915 to 1 000		8.10^{-8} $8.5.10^{-8}$	$6.2.10^{-8}$ $4.7.10^{-8}$
SL-P1	-	24.06.82	23.12.83	330	9.3 to 11.9	4.9 to 6.3	800	840	$5.8.10^{-7}$	$9.7.10^{-7}$	800	840	$1.2.10^{-6}$	$1.0.10^{-6}$
HFR-K3	1/A 2/B 3/C	15.04.82	05.09.83	359.3	7.5 10.0 10.2 9.0	4.0 5.8 5.9 4.9	950		$1.2.10^{-9}$ $9.1.10^{-10}$ $2.1.10^{-9}$	$1.2.10^{-9}$ 9.10^{-10} 2.10^{-9}	1 045 780 740 1 020		$2.1.10^{-7}$ $1.4.10^{-7}$ $2.5.10^{-7}$	$1.7.10^{-7}$ $1.6.10^{-7}$ $2.8.10^{-7}$
FRJ2-K13	1 2	24.06.82	12.02.84	396	7.5 8.0 7.9 7.6	< 10^{20}	970 1 020	Approx. 1 220	$2.0.10^{-9}$ $8.0.10^{-10}$	$1.5.10^{-9}$ 6.10^{-10}	970 970	Approx. 1 180 Approx. 1 180	$1.6.10^{-8}$ $6.6.10^{-9}$	$1.4.10^{-8}$ $5.4.10^{-9}$
FRJ2-P27	1 2 3	17.02.84	10.02.85	232.3	7.6 8.0 7.6	1.42 1.67 1.29		900 1 180 1 030	$4.2.10^{-6}$ $4.5.10^{-6}$ $2.0.10^{-8}$	$4.0.10^{-6}$ $4.0.10^{-6}$ $2.2.10^{-8}$		Approx. 900 Approx. 1 250 Approx. 1 100	$1.6.10^{-6}$ $4.5.10^{-6}$ $1.2.10^{-7}$	9.10^{-7} 4.10^{-6} 1.10^{-7}
FRJ2-P28	1 2 3	27.01.83	15.01.84	250.7	8.3 9.1 8.3	1.8 2.0 1.65		900 1 180 130	4.10^{-3} 7.10^{-3} 6.10^{-3}	$3.5.10^{-3}$ 6.10^{-3} 5.10^{-3}		900 1 150 1 010	$3.5.10^{-3}$ 6.10^{-3} $3.5.10^{-3}$	3.10^{-3} 5.10^{-3} $2.3.10^{-3}$

Explanation of Table 8 Column Headings:

The headings of the first two columns have exactly the same meaning as in **Table 5**.

a. Irradiation Start, End, and Duration

The irradiation duration in Full Power Days (FPD) is calculated by dividing the energy produced by the reactor during the residence time of the experiment (in MWd) by the nominal maximum power of the reactor (in MW).

b. Burn-up

Because of considerable inaccuracy in the calculations used to calculate burn-up during the experiments and to determine when to stop the irradiation, the values in the table are measured values determined during post-irradiation investigations by means of gamma spectroscopy and mass spectrometry.

For experiments HFR-P4/SL-P1, the burn-up range between maximum and minimum burn-up was specified. For the rest of the experiments, single values for the sample with the highest burn-up are given.

c. Fast Fluence E > 0,1 MeV

For experiments HFR-P4 and SL-P1, similar values to those specified for burn-up are shown.

d. BOL

BOL = Beginning of Life (start of irradiation).

Temperature °C: Surface temperatures were measured by capsule instrumentation. With the help of the calculated fission product gamma power of the test samples, the central fuel element temperature was estimated. The given values are averages taken over several days.

R/B (Kr 85m/Kr 88): A large number of measurements of fission gas release rates from irradiation capsules were performed using sweep gas loops available at a number of reactors. The release rates were specified in terms of R/B values (R = release rate; B = birth rate). In **Table 8**, average R/B values taken over the whole irradiation time (at least 20 FPD) for the isotopes Kr85m and Kr88 are shown. The release ra tes were determined from sweep gas measurements, taking into account sweep loop data such as geometry, pressure, and flow rate, while the birth rate was calculated as an equilibrium value determined by birth/decay data and the reactor power, taking into account operational and fuel data.

e. EOL

EOL = End of Life (end of irradiation).

Temperature °C: Probe power and the heat conductivity of the matrix determine the difference between the measured sample surface temperature and the temperature at the probe centre. As both sample power and heat conductivity depend on the thermal to fast neutron flux ratio, which varies for the individual reactors and irradiation positions, the relationship between surface and centre temperature was a complicated one, which changed as irradiation progressed.

In general the temperatures (surface and centre) stated in the table are those obtained from reliable measurements.

In the case of capsule C/03 of test HFR-P4, the temperature had to be estimated over a large part of the irradiation, because all thermocouples had failed. Estimates were based on reactor power, temperature and sweep gas composition.

R/B (Kr85m/Kr88): As a result of very low free uranium values for fresh fuel, fission gas release, expressed as R/B at BOL is very low (10^{-10} to 10^{-8}). The R/B ratio was expected to remain very low, providing that irradiation-induced particle failure did not occur. However, measurements showed a small steady increase in release rates during irradiation as shown in **Figure 2**. The figure shows the Kr88 release from capsule 1 of test HFR-P4 as function of irradiation time. Particle failure during irradiation could be excluded based on the very low R/B value at EOL, and the fact that no sudden increase occurred during the whole irradiation time. The increase in R/B values could be attributed to the known contamination of fuel matrix graphite with natural uranium and thorium. During irradiation, fissionable isotopes are produced from these materials through neutron absorption. Fission occurring in the fissionable isotopes caused the increase in fission gas release rates that are observed during irradiation tests.

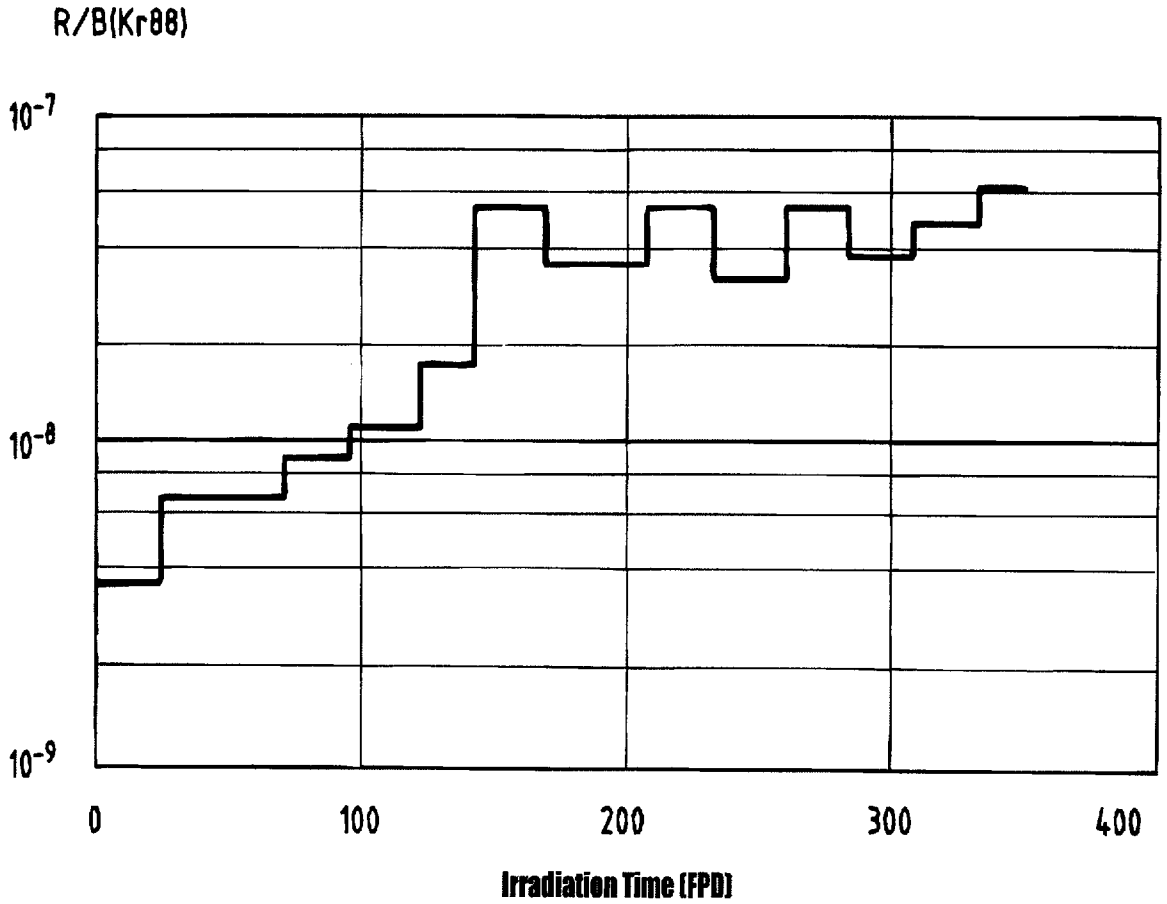


Figure 2: Kr88 R/B as a Function of Irradiation Time

Table 9: Phase 1 LEU-TRISO Programme – Calculations from Irradiation Data

Experiment	Capsule	Nominal Fuel Temperature	R/B (Kr88) from Defective Particles	Number of Particles in Capsule	R/B (Kr88) for One Defective Particle in Capsule (Calculated)	Measured R/B (Kr88)		Calculated Number of Defective Particles		
						BOL	EOL	BOL	EOL	Irradiation Induced
HFR-P4	A/01	1 000	7×10^{-3}	19 572	3.6×10^{-7}	3.6×10^{-9}	6.2×10^{-8}	0 (1×10^{-2})	0 (2×10^{-1})	0
	C/03	1 200	1×10^{-2}	19 572	5.1×10^{-7}	2.5×10^{-9}	4.7×10^{-8}	0 (5×10^{-3})	0 (9×10^{-2})	0
SL-P1	-	800	4.5×10^{-3}	19 572	2.3×10^{-7}	9.7×10^{-7}	1.0×10^{-6}	5 (4.22)	5 (4.35)	0
HFR-K3	1/A	1 200	1×10^{-2}	16 400	6.1×10^{-7}	1.2×10^{-9}	1.7×10^{-7}	0 (2×10^{-3})	0 (3×10^{-1})	0
	2/B	1 000	7×10^{-3}	32 800	2.1×10^{-7}	9×10^{-10}	1.6×10^{-7}	0 (4×10^{-3})	0 (8×10^{-1})	0
	3/C	1 200	1×10^{-2}	16 400	6.1×10^{-7}	2×10^{-9}	2.8×10^{-7}	0 (3×10^{-3})	0 (5×10^{-1})	0
FRJ2-K13	1	Approx. 1200	1×10^{-2}	32 800	3.0×10^{-7}	1.5×10^{-9}	1.4×10^{-8}	0 (5×10^{-3})	0 (5×10^{-2})	0
	2	Approx. 1 200	1×10^{-2}	32 800	3.0×10^{-7}	6×10^{-10}	5.4×10^{-9}	0 (2×10^{-3})	0 (2×10^{-2})	0
FRJ2-P27	1	900	5.8×10^{-3}	7 340	7.9×10^{-7}	4×10^{-6}	9×10^{-7}	5 (5.06)	1 (1.14)?	0
	2	1 300	1.3×10^{-2}	7 340	1.8×10^{-6}	4×10^{-6}	4×10^{-6}	3 (2.22)	3 (2.22)	0
	3	1 100	8.5×10^{-3}	7 340	1.2×10^{-6}	2×10^{-8}	1×10^{-7}	0 (1.7×10^{-2})	0 (8.3×10^{-2})	0

Explanation of

Table 9: **Column Headings:**

Experiment, Capsule, and Nominal temperature headings are the same as for **Table 5** .

Number of Particles in Capsule heading is the same as for **Tab le 7**.

Measured R/B (Kr 88), BOL and EOL headings are the same as for **Table 8:** .

a. R/B (Kr88) from defective particles

Results from test FRJ-P28, **Figure 3**, which contained a known number of defective particles, made it possible to calculate the R/B (for Kr88) value that would result if all particles were defective at the specified temperature. Because of the weak dependence of R/B on temperature the nominal fuel temperature could be used neglecting local temperature differences and the difference between BOL and EOL temperatures.

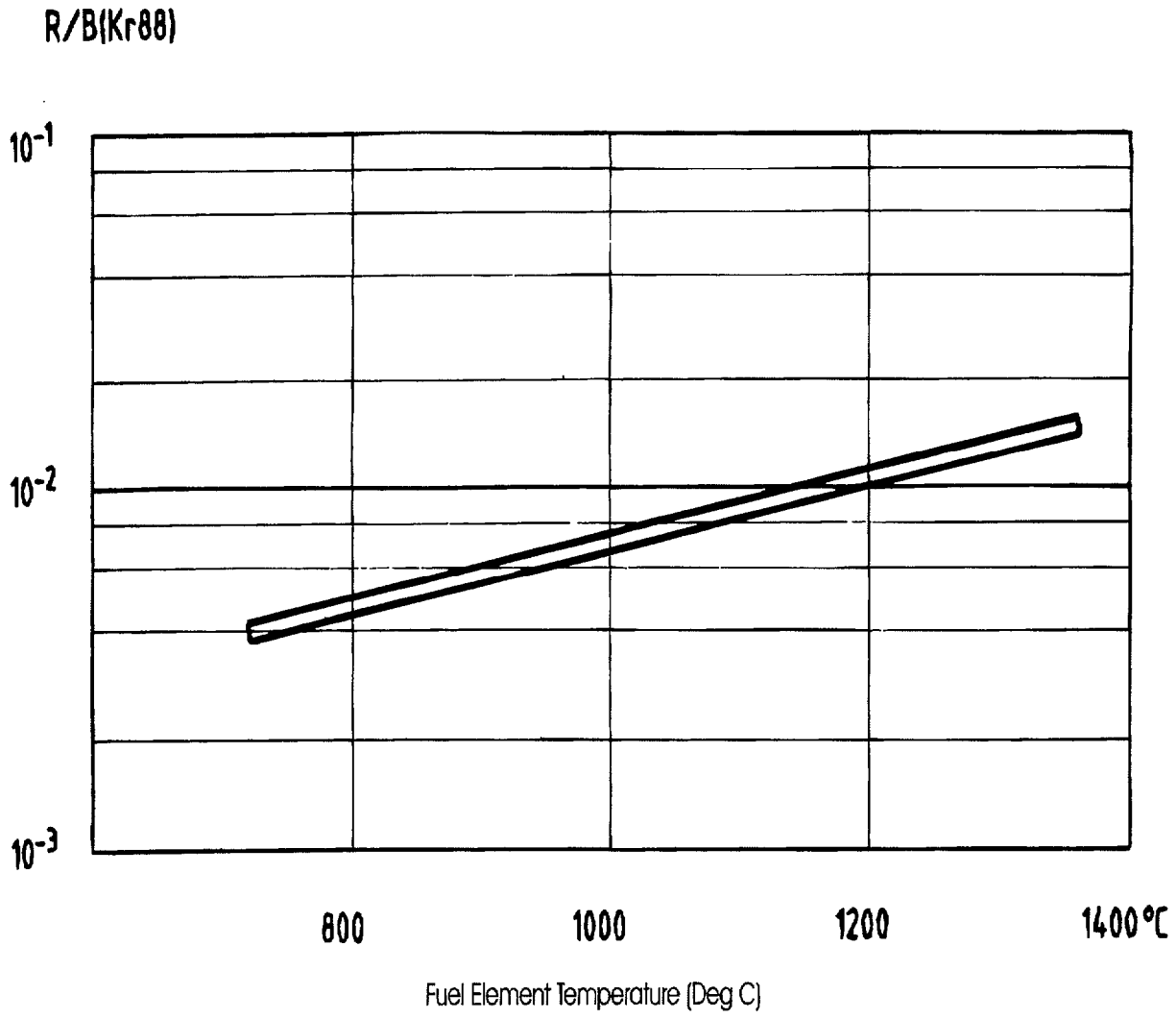


Figure 3: Kr88 R/B as Function of Fuel Element Temperature

b. R/B (Kr88) for 1 defective particle in capsule

An R/B value for a capsule containing a single defective particle could be calculated by multiplication of the R/B value from defective particles (above) with the inverse of the number of particles contained in the capsule. This calculation is valid provided that the release contribution from heavy metal contamination of the matrix is very low. The values in the column reflect the effects of temperature dependent release from defective particles, and the total number of particles contained in the capsule. If a sharp increase above this value is observed, it must be concluded that one or more particles have become defective.

c. Calculated number of defective particles

Division of the measured BOL and EOL R/B values by the calculated value for R/B resulting from a single defective particle, yields the number of defective particles in a capsule. If the resultant value is less than one and no sudden increase of the release rat e occurred for a capsule during irradiation, it means that all particles have remained intact. Calculated values are shown in brackets in the table, and the defective particle count is normally zero.

Five and three defective particles were found respectively in capsules 1 and 2 of test FRJ-P27, and for test SL-P1 5, defective particles were found at BOL.

At EOL, an identical number of defective particles was found in all cases, except for capsule 1 of test FRJ-P27. For this capsule, a smaller number (only one compared to five for BOL) was found, for which there is no explanation. Thus it could be concluded that no additional particles in any of the tests failed due to irradiation.

Phase 1 results showed a total number of 13 defective particles, leaving out experiment FRJ2 - P28, from a total of 211 936 irradiated particles. Combining this result with the three defective particles found in the quality control measurement on approximately 102 000 particles, a combined value of 5×10^{-5} for the free uranium fraction for manufactured fuel was measured.

In a similar way, the free uranium fraction for end of irradiation (EOL) conditions was determined. In none of the tests, excluding FRJ-P28 and AVR, was an increase in the number of defective particles found during irradiation. Thus none of the 211 936 particles irradiated failed during Phase 1 experiments, even under the extremely high irradiation conditions of test HFR-P4.

4.3.4 Post-irradiation investigations

The cumulative release fraction of Kr85 as a function of time for fuel elements from Phase 1 tests is shown in **Figure 4** for different annealing temperatures. The annealing was carried out at constant temperature, and the results are representative of full-size fuel elements with approximately 16 400 coated particles per element.

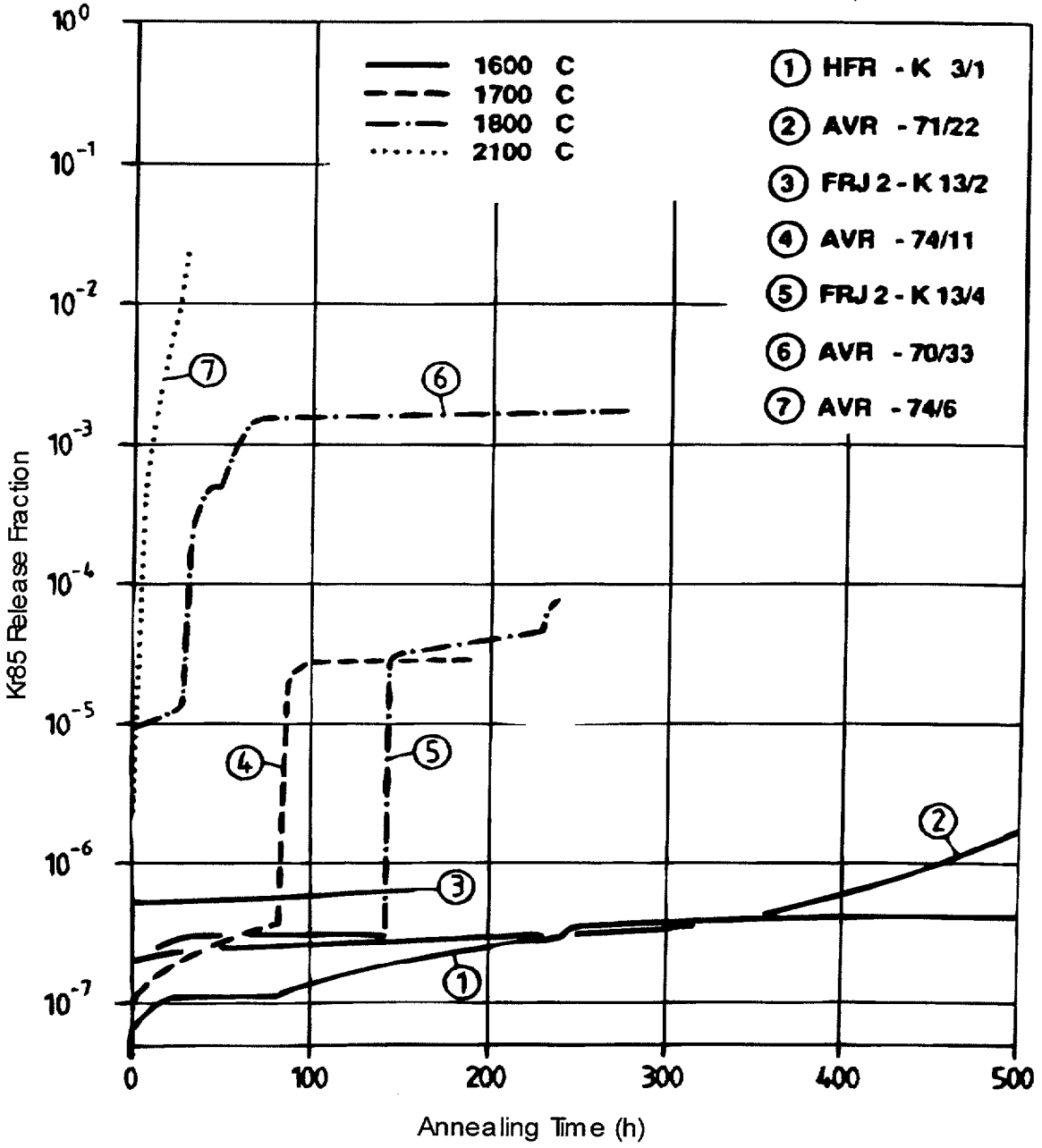


Figure 4: Kr85 Release Fraction as a Function of Annealing Time

The release fraction graphs for annealing at 1 600 °C (curves 1, 2, 3 and 5) show that fission gas release rates stayed at very low levels for long times (more than 100 h). This release was due mainly to the very low natural uranium contamination of the graphite matrix. It is also important to note that not a single particle failed during annealing of fuel elements. The temperature of fuel element number FRJ2-K13/4 (curve 5) was raised to 1 800 °C after being at 1 600 °C for 138 h and then annealed for a further 100 h at the higher temperature.

The broken curve designated as AVR 74/11 (curve 4) shows the results of annealing at 1 700 °C. The sudden rise in the curve after 85 h is characteristic of coated particle failure during annealing.

The release curve for annealing at 1 800 °C (curve 6) shows the effect of multiple coated particle failure (failure of SiC pressure vessel). The curve indicates the failure of approximately 40 coated particles in fuel element AVR 70/33. From annealing experiments on samples containing simulated defective particles from test FRJ2-P28, it became clear that the behaviour of iodine was similar to the behaviour of krypton and xenon. This means that iodine is almost completely contained within particles with intact coatings, and that the release of iodine is proportional to the defective particle fraction.

Results from annealing tests measuring the release of Cs137 and carried out on fuel elements similar to those used for Kr85 release measurements showed that the release pattern for Cs137 was similar to that of Kr85. However, the temperature dependence was not so marked as in the case of Kr85. The Cs137 release from failed particles is superimposed on the release due to thermally activated diffusion of Cs137 through intact SiC layers.

It is important to note that in all annealing tests carried out at 1 600 °C, not a single LEU-TRISO coated particle failed. On the other hand, it must be pointed out that the irradiation loads of the elements annealed at 1 600 °C did not simulate the burn-up target values for HTR-Modul very well. On the whole, the test burn-up values were lower. However, the fast fluence values for some of the tests were appreciably higher than the target values. One example was test element HFR-K3/1 with a fast fluence of 3.9×10^{21} n/cm^2, which was markedly higher than the target value for HTR-Modul. Refer to **Table 11** and **Table 13**.

The following observations were made from a study of ceramographic images of coated particles from different Phase 1 tests and annealing programmes:

- The appearance of the SiC layer of a coated particle from an irradiated fuel element (6.5%FIMA) was not much different from that of a coated particle from an unirradiated fuel element. It showed the same amorphous, dense white surface with very few small and dark, point-shaped pores.
- The SiC layer of a coated particle from a fuel element irradiated at 1 200 °C to a burn-up of 8% FIMA and annealed for 138 h at 1 600 °C was for all practical purposes the same as the SiC layer of an unirradiated coated particle.
- Some evidence of degradation of the inner surface of the SiC layer was visible for coated particles from a fuel element that had been irradiated to 3.5% FIMA and then annealed at 1 600 °C for 500 h.
- Under the same conditions (annealing at 1 600 °C for 500 h) the SiC layer for coated particles from a fuel element with increased burn-up and fast neutron fluence (8% FIMA at 1 200 °C; fast fluence 3.9×10^{21} n/cm^2) showed increased degradation. The decrease in density of the layer was much more visible.
- An element of average burn-up (6.2% FIMA) annealed at 1 700 °C for 185 h showed severe damage penetrating the SiC layer.
- Less severe, although penetrating, damage of the SiC layer was visible for a fuel element of low burn-up (1.8% FIMA) annealed at 2 000 °C for 100 h.
- Severe penetrating damage of the SiC layer was visible for a fuel element of average burn-up (5.6% FIMA) annealed for 30 h at 2 100 °C.

The following qualitative conclusions were drawn from the ceramographic investigations:

- The SiC layer of coated fuel particles and thus the fission product retention capability of fuel elements would remain intact under all foreseen normal operating conditions, and also
-

- during upset conditions leading to fuel element heating for long time spans.
- Even at 1 600 °C, heating for extremely long time spans (500 h) would result in damage to coated particles in fuel elements.
- Changes in the fission product retention capability of coated particles became apparent, even at 1 600 °C, when the irradiation load was very high, i.e. irradiation temperatures of 1 200 °C and fast neutron fluence of 4 x 10^{21} n/cm^2 or more.
- It was not clear which of the mechanisms burn-up, fast neutron fluence or fuel element temperature was dominant in the degradation of the SiC layer at temperatures of 1 600 °C and higher.
- At annealing temperatures of 1 700 °C and higher for relevant time spans, SiC damage must be expected.

Annealing tests on full-size fuel elements from Phase 1 experiments were supplemented with annealing tests on small spheres from HFR-P4 and SL-P1. The irradiation loads on these tests spanned the expected values for HTR-Modul a lot better than the other tests. The following preliminary results were obtained:

- Small spheres from HFR-P4 were irradiated to a burn-up of 11.1% FIMA, a fast neutron fluence of 5.5 x 10^{21} n/cm^2, and a temperature of 1 000 °C, and then annealed at 1 600 °C. The Kr85 release was very similar to curves 1 and 2 in **Figure 4**, i.e. it remained practically constant between 10^{-7} and 10^{-6} for up to 304 h.
- Small spheres from SL-P1 were irradiated to a burn-up of 10.7% FIMA, a fast neutron fluence of 6.7 x 10^{21} n/cm^2, and a temperature of 800 °C, and annealed at 1 600 °C. Very similar results to the above were obtained.
- A small sphere HFR-P4/3.7 was irradiated to a burn-up of 13.9% FIMA, a fast neutron fluence of 7.5 x 10^{21} n/cm^2 and a temperature of 1 200 °C, and then annealed at 1 600 °C. After 50 h, the first coated particles failed and the Kr85 release fraction started rising, reaching a value of 10^{-3} at 304 h.
- A small sphere (number 1.8) was irradiated to a burn-up of 13.8% FIMA, a fast neutron fluence of 7.2 x 10^{21} n/cm^2 and a temperature of 1 000 °C (halfway between the previous two temperatures), and then annealed at 1 600 °C. After 70 h, the first coated particles failed and the Kr85 release fraction started rising, reaching a value of 5 x 10^{-5} at 304 h.
- Caesium release was clearly higher for these samples. The Cs137 release fractions were spread between 3 x 10^{-4} and 4 x 10^{-3}. This was expected because caesium, unlike fission gases and iodine, is not contained by PyC layers at elevated temperatures.

The irradiation loads of all small spheres that were annealed were more severe than that expected for HTR-Modul.

Derivation of Base Data for the Radiological Design of HTR-Modul

The most important source for the release of fission products of radiological importance for HTR-Modul during normal operation and for upset event conditions was 'free uranium' or 'failed particle fraction' in the fuel elements. In order to quantify free uranium/failed particle fraction, the following three fuel conditions were distinguished:

- Initial condition (BOL): Failed particle fraction due to manufacture.
- Normal operation (up to EOL): Failed particle fraction induced by radiation.
- Core heating upset events (EOL): Failed particle fraction induced by high fuel temperatures.

4.3.5 Failed particle fraction for as-manufactured fuel

In the fuel specification for HTR-Modul, the average value for the free uranium fraction as determined by the burn-leach method for a lot consisting of 10 000 fuel elements was specified as 6×10^{-5}. This value was used as the design value for HTR-Modul fission product release from fresh fuel. A single defective particle in an HTR-Modul fuel element (11 600 coated particles per fuel element) would represent a failed particle fraction of 8.6×10^{-5}. Thus the specified failed particle fraction corresponded to 0.7 failed particles per HTR-Modul fuel element.

The actual free uranium values for the partial AVR loading AVR-19, 21/1 and 21/2 fluctuated between the detection limit for the burn-leach method of 1×10^{-5} and a maximum value of 5×10^{-5}. An expected value of 3×10^{-5} was accepted for LEU-TRISO fuel elements.

4.3.6 Radiation-induced failed particle fraction

Leaving out experiment FRJ2-P28 where failed particles were artificially introduced, the continuous monitoring of fission gas release during irradiation for all other experiments indicated that not a single particle of the 212 000 particles irradiated, failed during irradiation.

In applying the experimental result of 'no particle failure for 212 000 particles tested' to finding a design value for failed particle fraction at EOL for HTR-Modul, the following was considered:

- In Phase 1 experiments, coated particles from a single coating charge were used, while in the HTR-Modul equilibrium core, particles from approximately 250 to 500 coating charges would be present in the reactor at any one time. The uncertainty introduced by this factor was not considered to be very large in the light of experience with fuel elements from charge AVR-19, which contained coated particles from 65 coating charges.
- During reactor operational testing of LEU-TRISO fuel elements in the AVR, nine LEU-TRISO fuel elements from load AVR-19 were removed from the reactor and annealed at different temperatures. The specifications for these fuel elements are shown in **Table 10** and compared to specifications for Phase 1 test fuel elements.
- In order to simulate fission product release during normal operational conditions, annealing was done in three steps. Fuel elements were first heated at a temperature of 1 050 °C for several hours, and then at 1 250 °C for a further period of several hours before heating to the final annealing temperature. This procedure produced a fission product release rate that was equivalent to the release rate that would occur in an equilibrium situation in a reactor that operated at the temperatures involved. No evidence indicating an appreciable failed particle fraction is evident from the results shown in **Table 11**, **Table 12** and **Table 13**.
- Phase 1 experiments were performed in several different reactors, and the Modul target burn-up was not fully reached in some of the experiments, while they were exceeded in others. The same is true for the fast neutron fluence values. Thus all coated particles irradiated in Phase were taken as a single batch in calculating a radiation-induced failed particle fraction.
- All Phase 1 experiments, except FRJ-P28, were performed at constant temperature and power that were mostly higher than the expected values for HTR-Modul. In HTR-Modul, fuel elements were to be circulated 15 times (maximum 17 times) through the core during their lifetime, undergoing continuous changes in temperature and power in the process. This fact was not considered to contribute much to the uncertainty of applying Phase 1 results to HTR-Modul. The already-mentioned AVR annealing tests were performed on fuel elements that had undergone such cycles, although not as many cycles as in HTR-Modul.

Table 10: Annealing Tests LEU-TRISO – Comparison of Reference Fuel Elements and Fuel Elements from AVR-19

Parameter	Irradiation Experiment	
	AVR 19	HFR-K3/FRJ2-K13
Particle lot	HT 232-245	EUO 2308
Kernel composition	UO_2	UO_2
Kernel diameter (μm)	500 ± 2%	497 ± 3%
Kernel density (g/cm^3)	10.80	10.81
Coating Thickness (μm)		
Buffer layer	93 ± 14%	94 ± 11%
Inner PyC layer	38 ± 10%	41 ± 10%
SiC layer	35 ± 6%	36 ± 5%
Outer PyC layer	40 ± 9%	40 ± 6%
Coating Density (g/cm^3)		
Buffer layer	1.01	1.00
Inner PyC layer	1.86	No value
SiC layer	3.19	3.20
Outer PyC layer	1.89	1.88
Sphere identification	AVR-GLE 3	HFR-K3/FRJ2-K13
Type of matrix material	NUKEM A3-27	
Fuel Loading		
U235 (g/FE)	1	1
Heavy metal (g/FE)	10	10
U235 enrichment (%)	9.82	9.82
Number of particles per FE	16 400	16 400
Free Uranium Fraction (Burn-leach measurement)		
Particles	No value	$< 5 \times 10^{-8}$
Fuel element	5×10^{-5}	3.5×10^{-5}
Year of manufacture	1981	1981

Table 11: Annealing Tests LEU-TRISO - Sample Irradiation Data

Fuel Element	Burn-up (% FIMA)	Neutron Fluence (E > 0,1 MeV) x 10^{25} m^{-2} (a)	Full Power Days (a)	Average Irradiation Temperature (°C) (b)
FRJ2-K13/2	8.0	0.1	396	1 000 to 1 200
FRJ2-K13/4	7.6	0.1	396	1 000 to 1 200
HFR-K3/1	7.7	3.9	359	1 000 to 1 200
HFR-K3/3	10.2	6	359	800 to 1 000
AVR 70/33	1.6	0.2	210	~ 700
AVR 71/7	1.8	0.2	240	~ 700
AVR 70/19	2.2	0.3	290	~ 700
AVR 74/8	2.9	0.4	390	~ 700
AVR 73/12	3.1	0.4	410	~ 700
AVR 71/22	3.5	0.5	470	~ 700
AVR 74/10	5.5	0.8	740	~ 700
AVR 74/6	5.6	0.8	750	~ 700
AVR 74/11	6.2	0.9	830	~ 700
Modul	9	2.1	1 000 (c)	Max. 800
HTR 500	10.8	3.3	700 (d)	Max. 880

Notes:

a) AVR value obtained from interpolation of calculated values.

b) AVR temperature of 700 °C is an estimate – fuel centre temperatures varied from 300 to 1 150 °C

c) After multiple passes through the reactor core.

d) Single pass through the reactor core.

Explanation of Table 11 Column Headings:

In this table, the most important irradiation data for all LEU-TRISO samples annealed up to October 1986 are shown in comparison to design values for HTR- Modul and HTR 500.

The small deviations regarding the HFR-K3 and FRJ2-K13 values were due to the fact that their burn-up values could only be determined during post-irradiation investigations.

The burn-up values for the AVR elements were also determined in the hot cells at Jülich during post-irradiation investigation through Cs137 concentration measurements.

The irradiation time as well as the fast neutron fluence could not be determined exactly for single AVR elements. These values were calculated using nuclear physical relationships and AVR operational data.

Table 12: Annealing Tests LEU-TRISO - Results from Operational Test Phase

Fuel Element	Burn-up (% FIMA)	Annealing Time (h)		R/N Kr85 (s^{-1})		R/N Xe133 (s^{-1}) at End of Irradiation
		1 050 °C	1 250 °C	1 050 °C	1 250 °C	
FRJ2-K13/2	8.0	23	21	$< 1.2 \times 10^{-12}$	$< 1.2 \times 10^{-12}$	2.8×10^{-14}
FRJ2-K13/4	7.6	7	9	No equilibrium release	No equilibrium release	
HFR-K3/1	7.7	5	16.5	$< 6.8 \times 10^{-13}$	6.8×10^{-13}	6.1×10^{-13}
HFR-K3/3	10.2	5	13.5	3.5×10^{-12}	8.8×10^{-12}	3.0×10^{-13}
AVR 70/33	1.6	5	8	4.2×10^{-11}	7.0×10^{-11}	-
AVR 71/7	1.8	10	7	$< 2.2 \times 10^{-11}$	$< 2.2 \times 10^{-12}$	-
AVR 70/19	2.2	8	10	$< 5.2 \times 10^{-11}$	$< 5.2 \times 10^{-11}$	-
AVR 74/8	2.9	7	17	$< 4.0 \times 10^{-12}$	$< 4.0 \times 10^{-12}$	-
AVR 73/12	3.1	10	8	$< 4.5 \times 10^{-12}$	$< 4.5 \times 10^{-12}$	-
AVR 71/22	3.5	15	23	$< 1.2 \times 10^{-12}$	1.2×10^{-12}	-
AVR 74/10	5.5	7	9	$< 2 \times 10^{-11}$	$< 2 \times 10^{-11}$	-
AVR 74/6	5.6	10	6	$< 2.4 \times 10^{-12}$	$< 2.4 \times 10^{-12}$	-
AVR 74/11	6.2	7	8	No equilibrium release	No equilibrium release	-

Table 13: Annealing Tests LEU-TRISO – Results

Fuel Element	Burn-up (% FIMA)	Annealing Programme			Fraction Released				
		Temper-ature (°C)	1 250...T (h)	Time at T (h)	Kr85	Cs134	Cs137	Sr90*	Ag110m*
FRJ2-K13/2	8.0	1 600	7.5	138(160)	6.4E-7	1.0E-4	3.9E-5	3.8E-6	2.8E-3
HFR-K3/1	7.7	1 600	9	500	1.8E-6	1.3E-4	1.1E-4	8.3E-6	2.7E-2
AVR 71/22	3.5	1 600	7.5	500	4.0E-7	6.9E-5	2.0E-5	5.3E-6	9.0E-4
AVR 74/11	6.2	1 700	9	184.5	3.0E-5	8.4E-5	7.6E-5	8.3E-5	3.2E-2
FRJ2-K13/4	7.6	1 600 1 800	7.5 2	138 100	3.0E-7 7.2E-5	5.7E-6 9.7E-3	2.5E-6 9.9E-3		1.0E-3 1.0
HFR-K3/3	10.2	1 800	12	25	1.1E-5	1.2E-3	1.0E-3	3.6E-5	8.8E-2
AVR 74/10	5.5	1 800 1 800 1 800 1 800	14 12 12	30 30 30 90	1.3E-4 8.3E-4 8.3E-4 1.8E-3	<1.0E-2 5.0E-2 3.5E-2 8.5E-2	<1.0E-2 3.7E-2 4.2E-2 7.9E-2		
AVR 70/33	1.6	1 800 1 800 1 800 1 800	14 3.5 3.5	50 24.5 100 174.5	5.1E-4 9.9E-4 2.2E-4 1.7E-3	<1.0E-2 <1.0E-2 LOST	<1.0E-2 <1.0E-2 2.2E-2 2.2E-2		

Fuel Element	Burn-up (% FIMA)	Annealing Programme			Fraction Released				
		Temper-ature (°C)	1 250...T (h)	Time at T (h)	Kr85	Cs134	Cs137	Sr90*	Ag110m*
AVR 73/12	3.1	1 900 1 900 1 900	15 15	50 50 100	1.6E-5 1.2E-4 1.4E-4	<1.0E-2	<1.0E-2		
AVR 71/7	1.8	2 000 2 000	18 18	50 50 100	2.5E-6 8.3E-5 8.6E-5	LOST	5.0E-2 4.2E-2 9.2E-2		
AVR 74/6	5.6	2 100	18	30	2.4E-2	5.0E-1	4.7E-1		
AVR 70/19	2.2	2 400	27	-	1.0E-2	3.1E-2	3.0E-2		
AVR 74/8	2.9	2 500	27	-	4.6E-2	2.5E-1	2.5E-1		

* Only measured for annealing measurements performed in KÜFA apparatus.

R/N Values in Table 12:

R/N values in the table are calculated as follows. The measured relative Kr 85 release during a heating period is corrected for decay to the end of irradiation. This corrected relative release is then divided by the heating time in seconds to provide an average release rate.

Explanation of Table 13 Column Headings:

For all annealing experiments completed by October 1986, the annealing programme and results are shown in the table. Whereas the release of Cs134, Cs137, Sr90, and Ag110m could be measured very accurately in the 'KÜFA' apparatus, only relatively coarse measurements (1 to 2% accuracy) for Cs134 and Cs137 release could be performed in the 'A-Test' facility through gamma spectrometric determination of inventory loss.

Although 212 000 particles, which is a large but finite sample, were irradiated without a single failure, the expected value calculated from this result would be a value for a finite sample. In the light of the above-mentioned qualitative simplifications, the expected value for the failed particle fraction due to irradiation was calculated for a 95% confidence value instead of the usual 50% confidence level. The expected failed particle fraction value found from statistical considerations **[10]** was 2×10^{-5}.

From this conservatively calculated expected value for the failed particle fraction, the design value was calculated by multiplying the expected value by a factor 10. Thus the design value for the failed particle fraction due to fuel irradiation under normal operating conditions for HTR-Modul was 2×10^{-4}.

Considering the fact that the HTR-Modul target values for burn-up and fast neutron fluence had not been achieved in some of the experiments, the following was stated. In experiments HFR-P4 and SL-P1 as well as spheres 2, 3, and 4 from HFR-K3, the HTR- Modul target values were achieved and even exceeded. Approximately 108 000 particles were irradiated in these experiments. Thus the 95% confidence interval expected value for the failed particle fraction, calculated from the observation that not one of these particles failed during irradiation, was 4×10^{-5}. This value could be used as a design value, in which case it would be a much better value than the very conservative design value of 2×10^{-4} found by including all irradiated particles.

However, taking 4×10^{-5} as an expected value as before, and using the same conservatism as before, a design value for the failed particle fraction could then be calculated as 4×10^{-4}, which was just a factor 2 more than the actual design value used for HTR -Modul.

The expected and design values for radiation-induced failed particle fraction for HTR- Modul, 2×10^{-5} and 2×10^{-4} respectively, were valid for fuel elements that have been irradiated to maximum burn-up. No relationship between burn- up and failed particle fraction could be distinguished from the results of Phase 1 experiments. Thus for HTR- Modul design, the conservative assumption was made that the irradiation-induced failed particle fraction increased linearly with fuel burn-up, starting at zero for fresh fuel and reaching the above value at maximum burn-up. The design value for HTR- Modul was fixed at a point halfway between zero and 2×10^{-4}, i.e. at 1×10^{-4}.

4.3.7 Failed particle fraction due to upset event temperature effects

The results of annealing tests at 1 600 °C for the following experiments were used as basis for the determination of a design value for temperature-induced failed particle fraction for HTR-Modul:

- FRJ2-K13 spheres 2 and 4 (8% and 7.6% FIMA respectively).
- HFR-K3 sphere 1 (7.5% FIMA).
- AVR fuel element 71/22 (3.5% FIMA).

No irreversible increase in fission gas release was noted during annealing at 1 600 °C of any one of these fuel elements. Refer to **Table 13** for AVR results. In the same way as described above, an expected value for temperature-induced failed particle fraction of 5×10^{-5} at 95% confidence level was derived, taking into account the number of coated particles contained in the fuel elements.

Using the same conservatism as above, a design value of 5×10^{-4} for temperature-induced failed particle fraction was found. This value was used for the volume fraction of the HTR-Modul equilibrium core that would reach a temperature of 1 600 °C during an offset event. The design value of 5×10^{-4} at 1 600 °C was valid for all burn- up and fast neutron fluence values occurring between loading and unloading of the fuel. There was no evidence of any relationship between upset event temperature-induced particle failure and HTR-Modul operational conditions such as burn-up, fast neutron fluence, fuel temperature, and upset event course. In fact, it was clear from annealing tests on experiments HFR-P4 and SL-P1 that even for irradiation loads far beyond HTR-Modul limiting loads, the temperature -induced particle failure fraction was very low.

In three cases, the experiments used to determine the design value for temperature -induced failed particle fraction did not reach HTR- Modul target burn-up values. The AVR fuel element with a burn-up of 3.5% FIMA was clearly well below the target value. Fuel element 1 from HFR-K3, with a fast neutron fluence of 3.95×10^{21} n/cm^2, exceeded the targe t value of 2.1×10^{21} n/cm^2 by far. As mentioned before, the design value for temperature-induced failed particle fraction was valid for all HTR-Modul load conditions. The volume fraction of the core that would reach a temperature of 1 600 °C was very small, and the fraction of fuel elements that would have burn-up values exceeding the test values was even smaller.

Thus the conservatism (10 times the 95% confidence interval value) in the temperature -induced failed particle fraction would take account of the statistical uncertainties discussed above. This

was confirmed by the fact that annealing tests at 1 600 °C on samples from experiments HFR-P4 and SL-P1 showed the onset of particle failure only at extremely high irradiation loads.

During an upset event, there is a temperature distribution among the fuel elements in a reactor core. Fuel element temperatures will vary between the normal operating temperature and the maximum upset event temperature of 1 600 °C. As no particle defects were detected at normal operation conditions as well as under upset event conditions, some conservative but realistic assumptions were called for. It was assumed that particle failure due to temperature effects commenced at a temperature of 1 200 °C. The basis for this assumption was the fact that no particle defects were detected for Phase 1 experiments irradiated at temperatures of 1 200 °C. Furthermore, no particle defects were found in the fission gas release studies from LEU-TRISO fuel elements annealed at 1 250 °C. It was assumed that the mean failed particle fraction varied exponentially between 'no temperature-induced particle failure at 1 200 °C' and the design value of 5×10^{-4} at 1 600 °C. For the purposes of HTR- Modul design calculations, it was assumed conservatively that the failed particle fraction increased instantly with an increase in temperature, thereby excluding any failure mechanism that might be time dependent.

4.4 Conclusion

The HTR-Modul was thus designed in such a way that fission product release remained low and calculable under all operating conditions, and that any additional and unquantifiable mechanisms that would influence the retention capability of fuel for fission products could be excluded under all normal and upset event conditions.

The experimental evidence confirmed fuel quality regarding the sufficiency and calculability of fission product retention under normal and upset conditions for HTR- Modul and provided proven values for the amount of uranium not covered by intact SiC layers under different operating conditions investigated. This evidence consisted of experiments on appropriate fuel elements using experience gained from previous irradiation experiments, operational experience on AVR, and Phase 1 irradiation experiments as well as high temperature annealing experiments on irradiated fuel elements.

From the evidence, the following conclusions regarding normal and upset event operating conditions were made.

4.4.1 Normal operation

Fission product release from fuel elements in HTR-Modul under normal operating conditions could be attributed to two main causes:

- free uranium resulting from manufacturing processes; and
- irradiation-induced failure of coated particles.

Free uranium fraction (failed particle fraction) resulting from manufacture was specified as 6×10^{-5} per fuel element batch, and this value was used as design value for as-manufactured fuel elements. The expected value could be taken as 3×10^{-5}, and it was based on experience with the production of similar fuel elements for AVR. The free uranium in fresh fuel could be attributed almost entirely to single coated particles with defective SiC coatings. As the free uranium fraction due to a single defective coated particle in a fuel element amounted to approximately 8.6×10^{-5}, the design value of 6×10^{-5} corresponded to 0.7 failed particles per fuel element on average.

An irradiation-induced failed particle fraction could not be determined from Phase 1 experimental results. A conservative statistical calculation, taking into account the finite number of particles irradiated, was used to determine an expected value of 2 $x10^{-5}$ for the irradiation-induced failed particle fraction. This 95% confidence level value was multiplied by a factor 10 to find a design value for the irradiation-induced failed particle fraction. The factor 10 difference between expected and design values reflected the uncertainties in applying the results of Phase 1 experiments to HTR-Modul operational conditions.

These uncertainties stemmed from the fact that:

- Coated particles from only one batch were irradiated.
- One half of the irradiated coated particles did not reach HTR- Modul irradiation loads, while the other half exceeded Modul irradiation loads.
- Phase 1 irradiations were performed at nominally constant temperature and power, while fuel elements would be circulated through the HTR-Modul core a number of times, undergoing temperature and power cycles.

Based on AVR operational experience with similar LEU-TRISO fuel elements, these uncertainties were considered to be small, but th ey would be investigated further during Phase 2 irradiation experiments.

Using the above design values for the free uranium fraction of fresh fuel elements and for irradiation-induced failed particle fraction, an average and a maximum failed particle fraction of 1.6×10^{-4} and 2.6×10^{-4} respectively, was calculated for the Modul equilibrium core load of fuel elements that have reached full burn-up. These values corresponded to expected values of 4×10^{-5} and 5×10^{-5} respectively.

Compared to the release of fission products from defective coated particles, the release from intact particles was negligible.

4.4.2 Upset events resulting in core heat-up

A design value of 5×10^{-4} for temperature-induced failed particle fraction was deduced from the results of four fuel elements that were annealed at 1 600 °C for 100 h after being irradiated. This value was based on the result that no particle failure leading to an irreversible increase in iodine release was observed during annealing tests on these fuel elements. The design value was obtained by multiplying the 95% confidence level expected value of 5×10^{-5} by a factor 10. This design value was valid for the fraction of fuel elements of the Modul equilibrium core that would reach a temperature of 1 600 °C under upset event conditions.

The relatively large uncertainty reflected by the large difference between expected and design value was attributed to the fact that some of the fuel elements used in the tests did not reach the Modul target values for burn-up. Later experiments on LEU-TRISO fuel elements that have been irradiated to burn-up values far exceeding Modul values and then annealed, indicated that the temperature-induced failed particle fraction for Modul should not exceed the design value stated above.

A temperature of 1 200 °C was taken as the onset temperature for temperature -induced particle failure, and an exponential relationship between failed particle fraction and temperature was

assumed for the temperature range 1 200 °C to 1 600 °C. The conservative assumption was made that particle failure occurred without time delay once a certain temperature is reached.

Ceramographic investigations of irradiated LEU-TRISO fuel elements that have undergone annealing tests at 1 600 °C and higher temperatures were conducted. They indicated that the fission product retention capability of fuel elements irradiated at loads similar to those in HTR-Modul (9% FIMA burn-up, fast neutron fluence of 2.10×10^{21} n/cm^2 and maximum temperature of 865 °C) was not affected adversely in the sense of uncontrolled increase in the release of radiologically important fission products, irrespective of the temperature history of the upset event.

Changes in the SiC layer only became visible at irradiation loads that far exceed the upset even t loads and time spans expected for HTR-Modul. It could not be stated which of the factors – burn-up, fast neutron fluence, or irradiation temperature – was the dominant factor that caused the observed changes.

4.4.3 Summary

The failed particle fraction design values are shown graphically in **Figure 5** for the different load conditions for HTR-Modul fuel elements and operating temperature regimes. Graph number '2' describes the failed particle fraction calculated for a fuel element that has reached 50% of the target burn-up, using the assumption that failed particle fraction varies linearly with burn-up for the equilibrium core of HTR-Modul. Graph number '1' describes fuel that has reached the full target burn-up, while graph number '3' was used to describe fresh fuel. As described in **paragraph 4.3.7**, an exponential function was assumed between 1 200 °C and 1 600 °C for the HTR-Modul core loading. This function was independent of burn-up. The same absolute failure fraction values calculated for graph '2' were used to determine graphs '1' and '3' and this, plotted on a logarithmic scale, causes the slightly bent curves for the temperature dependent failed particle fraction in the latter graphs.

The calculations leading to **Figure 5** are summarized in **Table 14**.

Table 14: Summary of Calculated Values Used in Figure 5

Fuel Condition	Failed Particle Fraction		
	As Manufactured	As a Result of Irradiation	As a Result of Heating to 1 600 °C
BOL	6×10^{-5}	-	$6 \times 10^{-5} + (10)(5 \times 10^{-5}) = 5.6 \times 10^{-4}$
DESIGN	-	$(6 \times 10^{-5} + 2.6 \times 10^{-4})/2 = 1.6 \times 10^{-4}$	$1.6 \times 10^{-4} + 10(5 \times 10^{-5}) = 6.6 \times 10^{-4}$
EOL	-	$6 \times 10^{-5} + (10)(2 \times 10^{-5}) = 2.6 \times 10^{-4}$	$2.6 \times 10^{-4} + 10(5 \times 10^{-5}) = 7.6 \times 10^{-4}$

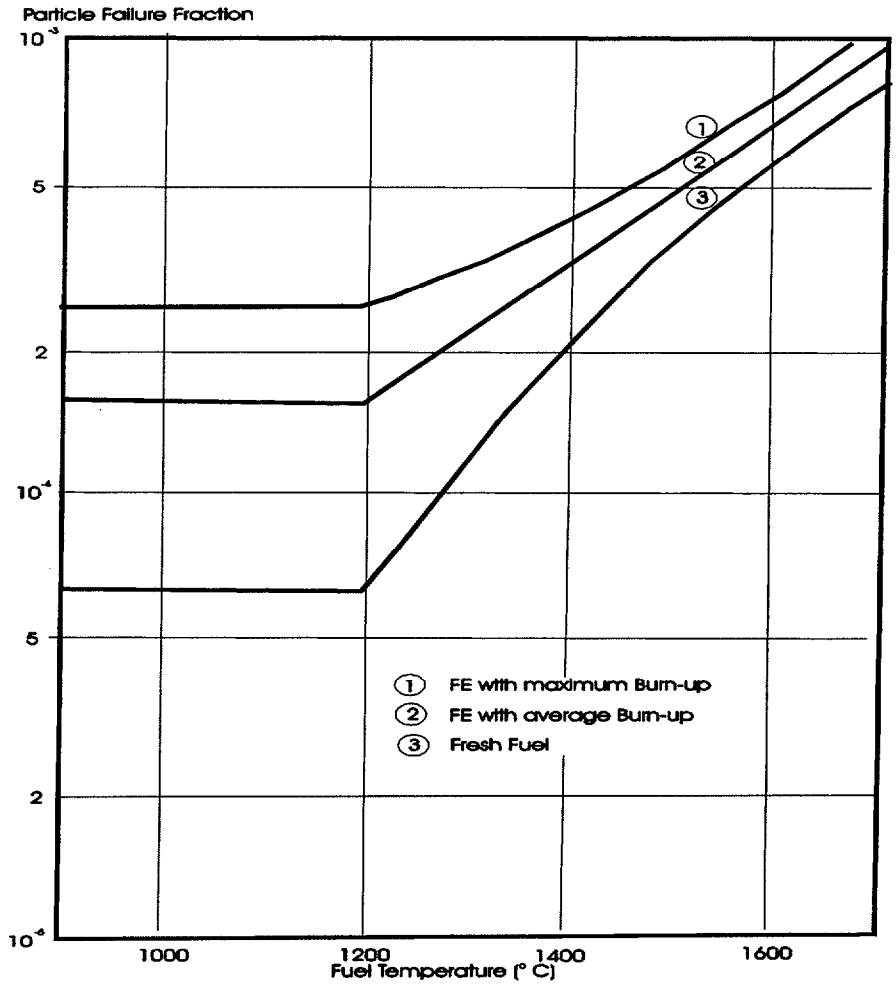


Figure 5: Failed Particle Fraction as Function of Fuel Temperature (Design Values)

The average failed particle fraction of 1.6×10^{-4} for the HTR- Modul equilibrium core contributes to the fission product release at all temperatures above normal operating temperatures. This release adds up to an appreciable fraction of the total release during core heat-up events. The temperature dependent failed particle fraction is not dominant in fission product release during upset events, because only about one-third of the fuel elements in the HTR-Modul core would reach temperatures higher than 1 200 °C, as can be seen from **Figure 6**. Only about 6% of the fuel elements would reach temperatures higher than 1 400 °C, where the failed particle fraction reaches a value of 3.3×10^{-4}, which is double the value for the equilibrium core at 1 200 °C. Thus the effects of uncertainties in failed particle fraction at elevated temperatures were relatively small.

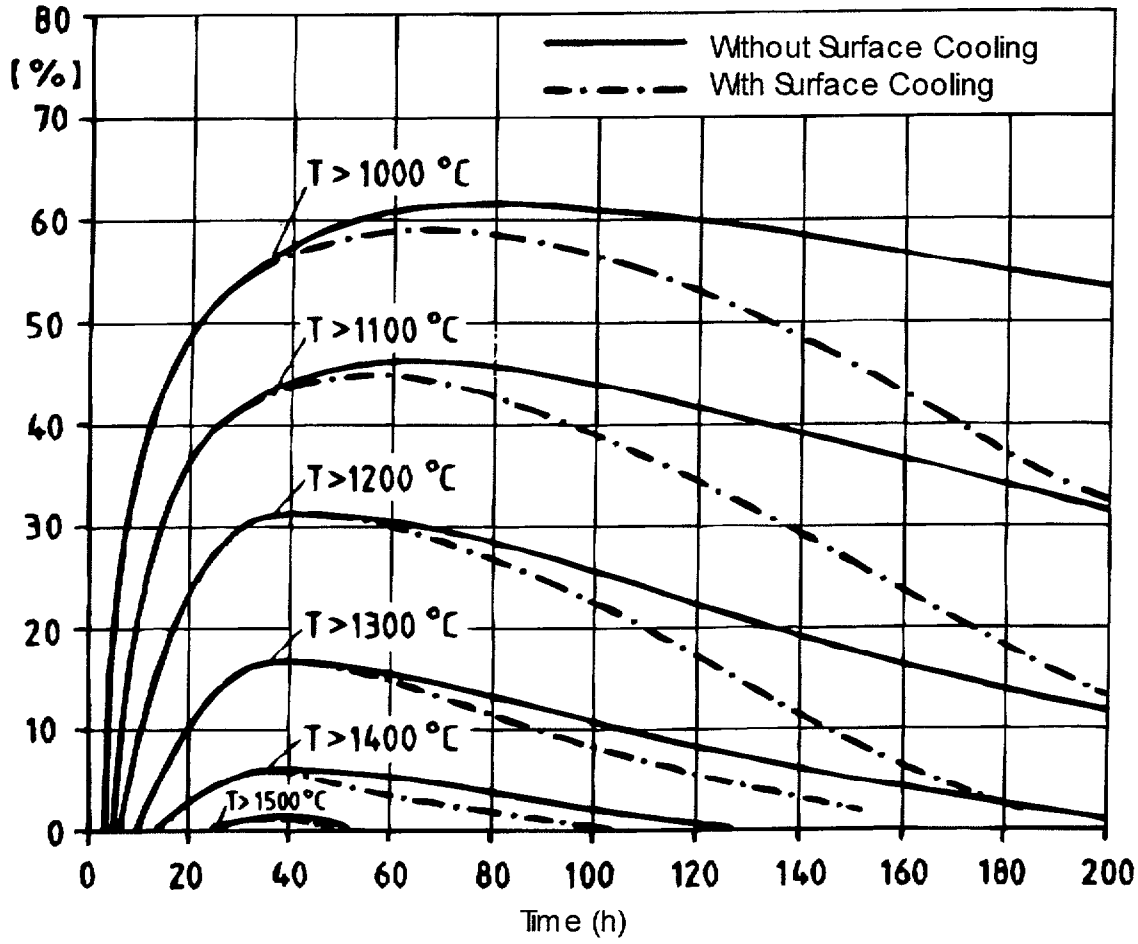


Figure 6: Volume Fraction of Reactor Core Above Specific Temperatures

The fission product release sources, deduced from Phase 1 experiments and AVR experience using similar fuel elements, contained a whole array of conservative arguments:

- In estimating the free uranium fraction of as- manufactured fuel, all uranium not covered by an intact SiC layer is included. Thus this fraction also contains uranium covered by intact pyrocarbon layers that have appreciable fission product retention capabilities for fission gases and iodine, even at temperatures as high as 1 600 °C.
- The 95% confidence level experimental values for radiation-induced and temperature - induced failed particle fraction were both multiplied by a factor of 10 to calculate design values.
- The assumption of a linear relationship between radiation-induced failed particle fraction and burn-up from BOL to EOL.
- Assumption that temperature-induced failure commences at a temperature of 1 200 °C.
- Assumption that particle failure follows spontaneously on an increase in temperature without any time delay.
- In determining temperature-induced failed particle fraction by means of annealing experiments, the temperature was kept at 1 600 °C for at least 100 h, which far exceeded HTR-Modul requirements.
-

The failed particle fraction values reflected the status of experimental results and the statistical significance of calculations based on these results for normal and upset event operational conditions for HTR-Modul.

The resulting fission product release sources calculated from failed particle fraction design values could be used for radiological design purposes, leading to sufficiently low activity burdens to personnel and the environment.

It was proven that irreversible and incalculable mechanisms that could damage the SiC layer of coated particles could be excluded if a maximum temperature of 1 600 °C for fuel elements was accepted. Taking 1 600 °C as the maximum allowable fuel temperature, other fuel element loads occurring simultaneously with temperature must also be considered.

5. IRRADIATION QUALIFICATION PROGRAMME: PHASE 2

5.1 Introduction

The description of Phase 2 tests given here is taken largely from **[3]** and **[4]**. The purpose of Phase 2 irradiation tests (Proof Tests) was to simulate as closely as possible the fuel element manufacturing process and HTR-Modul operational conditions to confirm the applicability and transferability of data from Phase1 irradiation tests to the Modul plant. Two reference irradiation tests, HFR-K5 and HFR-K6, with four test fuel elements each, were performed in the HFR at Petten in the Netherlands.

The latest manufacturing technique for coated particles was used to produce fuel for these tests. In this technique, approximately 40 kg of UO_2 kernels were manufactured in 22 batches and homogenized (kernel lot UOS 350). The homogenized kernels were coated in eight 5 kg lots (EUO 2358-2365) and characterized individually, and also as a combined lot. The lot was then coated in six runs and used to manufacture 150 fuel elements and 200 graphite spheres using the same matrix powder for both fuel elements and graphite spheres.

The maximum requirements placed on fuel elements in HTR- Modul were used to determine Phase 2 irradiation requirements, allowing some margin for uncertainties. The main test parameters are shown in **Table 15**, together with HTR-Modul requirements.

Table 15: Phase 2 Irradiation Targets

Property	Unit	HTR-Modul		Phase 2 Test
		Expected	Design	Target
Maximum Burn-up	% FIMA	8.9	9.8	10
Maximum Fast Neutron Fluence (E > 0.1 MeV)	10^{21} n/cm^2	2.13	2.4	2.4
Maximum Fuel Element Centre Temperature (Normal Operation)	°C	837	926	1 000
Maximum Fuel Element Centre Temperature (After heat removal via surface coolers on failure of main heat sink)	°C	-	1 130	1 200 (for 3 to 5 h)
Maximum Power per Fuel Element	kW	1.4	1.6	$2.0 \leq P \leq 3.6$
Maximum Power per Coated Particle	mW	130	150	$130 \leq p \leq 250$
Number of Core Passes	-	15	17	17
R/B (Kr88)	-	1.9×10^{-7}	6.1×10^{-7}	$< 6.1 \times 10^{-7}$

The most important characteristic of the irradiation programme was the simulation of temperature cycles encountered by fuel elements as they are cycled through the reactor core. In HTR-Modul, fuel elements would be cycled 15 times before being discarded. To allow for inaccuracy in the burn-up measurement, 17 temperature cycles were used in the irradiation tests. Fuel temperatures were kept at 800 ± 50 °C for one -third of the irradiation time, and at 1 000 ± 50 °C for two-thirds of the time. In order to simulate decay heat transients without loss of pressure, the test temperature was raised from 1 000 to 1 200 °C for 3 to 5 h three times during irradiation at the start, middle and end of irradiation.

Due to technical requirements of the reactor and the rig in which irradiation experiments were to be done, the enrichment and heavy metal loading for the test elements had to be changed from HTR-Modul values. For HTR-Modul, the enrichment was 7.7%, and the heavy metal loading was 7 g U per fuel element, which correspond to approximately 11 600 coated particles per fuel element. For the Proof Test, fuel of 10.6% enrichment was used with 9.435 g U per fuel element, i.e. 16 400 coated particles per fuel element. These adjustments were not considered to be critical for the test results.

With the fuel elements described in **Table 16**, the irradiation programme was completed in 634 days (26 HFR cycles) in the High Flux Reactor (HFR), despite the fact that the fission power in the fuel elements decreased appreciably during this time. The irradiation was performed in a position next to the HFR core, which provided the best thermal to fast neutron flux ratio for HTR-Modul irradiation relationships.

In the following paragraphs, the German philosophy for fuel irradiation tests will be discussed under the following headings:

- General purpose.
- Design base of fuel elements.
- Estimates of reactor irradiation conditions.
- Irradiation test.

5.2 Test Objectives

The qualification programme was required to provide the proof that LEU- TRISO fuel elements would perform according to HTR-Modul design specifications under all expected conditions in the reactor core, for the full expected design life of the fuel element. It was also required to provide the database for the fission product release source te rm during operation of the reactor.

The Proof Test for HTR-Modul combined reactor specific conditions for fuel elements and their irradiation requirements with post-irradiation procedures in a single experimental programme. It consisted of:

- HTR-Modul specific irradiation testing that would qualify fuel element design and specifications, as well as the materials used and the processes followed in the manufacturing of the fuel.
- The testing of fuel elements under nuclear and thermal conditions expected during normal HTR-Modul operation, as well as conditions expected during HTR- Modul upset event conditions, by simulating these conditions as closely as possible.

The Proof Tests were designed to provide the proof that material properties and changes in those properties, as determined in previous qualification tests, were transferable from simplified irradiation conditions to realistic conditions.

It was impossible to simulate all HTR-Modul conditions exactly in a single irradiation test. For instance, there was a difference in the enrichment and heavy metal loading between the test fuel elements and normal reactor fuel elements. This was due to the requirements imposed by the test reactor. In its normal lifetime, a fuel element would circulate through the reactor core 10 times, and there is no way to predict exactly where in the core this would happen each time.

Thus a single fuel element would experience a wide variety of neutron fluxes and temperatures during its life in the reactor. The design of the irradiation test required a fine balance in the choice of irradiation conditions involving the experimentalist, the fuel manufacturer and the reactor designer. This ensured an irradiation test that was representative of expected conditions in HTR-Modul.

The irradiation test was designed with the maximum requirements expected during operation with the equilibrium core of HTR-Modul in mind. Whenever the requirements for the burn -in core were higher than for the equilibrium core, allowance was made for this. Thus the fuel element power for the Proof Test was somewhat higher than expected for the equilibrium core. The temperature requirement for the irradiation test was also specified somewhat higher than expected in HTR-Modul operation, as will be explained later.

5.3 Description of the Irradiation Test

In the following paragraphs of the report, the selection of a set of minimum requirement parameters for the HTR Modul will be justified and described. The test will be described in terms of the design, manufacturing and characterization of the test sample on the one hand, and the design and development of the irradiation experiment itself on the other hand. This is done within the limits set by the reactor in which the irradiation was to be performed, keeping the above purpose statement in mind.

5.3.1 Test fuel elements

As explained above, it was necessary to reach a compromise between requirements and what was technically possible to achieve in an irradiation test. However, the compromise reached was not in any way to raise questions as to the achievement of the test objectives stated in ***paragraph 5.2***, and the set of minimum requirements was not to be compromised.

5.3.1.1 Fuel element design specifications

The specifications for Proof Test test fuel elements are set out in **Table 16**. In the table, the symbols used have the following meaning:

- x = parameter value
- [x] = average value
-

Table 16: Proof Test Fuel Element Design Specification – HTR Modul

Parameter	Unit	Design Specification
Fuel Kernel:		
Fuel Cycle		UO_2
Fuel Composition	UO_x	$x \leq 2.015$
U235 Enrichment	% U235	10.7 ± 0.1
Kernel Diameter	µm	$(450 - 550)_{95/95}$
Kernel Density	g/cm^3	$[x] \geq 10.4$ (≥ 95% theoretical density)

Parameter	Unit	Design Specification
Coated Particles:		
Buffer Layer:		
Thickness	µm	$(50 - 140)_{95/95}$
Density	g/cm^3	[x] ≤ 1.05
Inner Pyrocarbon Layer:		
Thickness	µm	$(25 - 55)_{95/95}$
Density	g/cm^3	[x] = 1.9 ± 0.1
Anisotropy (BAF)		[x] ≤ 1.10
Silicon Carbide Layer:		
Thickness	µm	$(25 - 45)_{95/95}$
Density	g/cm^3	[x] ≥ 3.18
Outer Pyrocarbon Layer:		
Thickness	µm	$(25 - 45)_{95/95}$
Density	g/cm^3	[x] = 1.9 ± 0.1
Anisotropy (BAF)		[x] ≤ 1.10
Fuel Element:		
Matrix material		A3 –3
Thickness of fuel free layer	mm	$(\geq 5)_{95}$/ $(\geq 4.5)_{100}$
Matrix density	g/cm^3	[x] = 1.75 ± 0.02
Heat Conduction Coefficient (1 000 °C)	W/(m.K)	[x] ≥ 25
Standard Corrosion Rate (1 000 °C)	mg/(cm^2.h)	[x] ≤ 1.3 x ≤ 1.5
Thermal Expansion Anisotropy		[x] ≤ 1.3
Breaking Strength	kN	x ≥ 18
Fall Resistance (4 m onto pebble bed)	Number of Falls	x ≥ 50
Abrasion Resistance 100 h in rotating drum	mg/h	[x] ≤ 6
Heavy Metal Content	g U/element	9.435
U235 enrichment	%U235	10.70 ± 0.1
Defective Particle Fraction (Burn leach method)		[x] ≤ 6 x 10^{-5}
Impurities – Boron Equivalent	ppm B	[x] ≤ 1.3

The following two parameters were changed for the irradiation test, due to requirements imposed by the HFR reactor used for the irradiation programme.

5.3.1.1.1 U235 enrichment

In order to reach the required burn-up and fast neutron dose in the test reactor simultaneously, it was required to use an enrichment of 10.60%, which is slightly higher than the 7.7% specified for Modul fuel. However, the maximum allowable enrichment was 10.7%, which was the upper limit that would allow 10 kg charges of UO_2 to be used in future fuel production.

5.3.1.1.2 Heavy metal content per fuel element

The heavy metal content was increased from 7.09 g for HTR-Modul to 9.435 g per fuel element for test fuel elements.

5.3.1.2 Fuel manufacture

The test objectives stated above required that the manufacturing process for test fuel elements and production fuel elements be equivalent. This objective had a bearing on the foreseen introduction of 10 kg coating charges for future fuel production.

In order to produce larger batches, it was foreseen that more than one coating charge would be combined and homogenized to form a coated particle batch. Although this process would increase the statistical weight of test results, it would yield no additional technical advantage.

Experience gained from statistical analyses has shown that for TRISO particles, all arguments were centred on zero values as far as irradiation-induced failure is concerned. This required dealing with purely calculated effects that added no technical/practical advantage. If particles failed due to the homogenizing process (which is highly improbable), it would be impossible to determine the causes of such, presently unknown, batch size effects on the strength of statistical evidence alone.

5.3.1.3 Characterization

Irradiation testing of fuel elements was done according to a sample design (target specification) and characterization programme. It specified all measurements that would be performed on the test sample (coated particles, matrix, and fuel elements), as well as the scope of the investigations. The programme specified standard quality, irradiation rigs and quality control procedures that were normally applicable, as well as additional requirements determined by the test target. It specified individual measurements and investigations that would be performed to reach the objectives of the irradiation test, and it also described the most important data that influenced the irradiation behaviour of the fuel elements being tested.

It was also important to describe important characteristics of materials, which fell outside the scope of the test, but had a definite influence on the test results. An example is the quality of the SiC layer of coated particles. It was known that the SiC structure influences the fission product retention of SiC. Therefore it was important to characterize the SiC structure during testing by means of ceramography and electron microscopy, and to document the results.

It was foreseen that it would be important that coated particles of Proof Test Standard Quality and a sufficient number of coating charges from future production charges be irradiated to prove that the fission product retention capability of intact coated particles is independent of charge size. This was to be done by proving that the diffusion constants lie within, or were better than

the HRB Standard Data Set distribution. Doing this would prove that the retention capability of intact coated particles, as specified through the manufacturing procedure for SiC, was sufficient, as this property could not be specified directly in terms of measurable material properties.

5.3.2 Irradiation

The second main objective of the irradiation test was to provide a realistic irradiation programme to simulate irradiation conditions expected in the HTR-Modul. This involved defining a list of requirements and the design of an experiment that would satisfy these requirements.

5.3.2.1 Technical requirements

In previous irradiation experiments, two irradiation capsule types had been developed:

- Two capsules containing two fuel elements each.
- Three capsules with two fuel elements in the middle capsule and one fuel element in each of the outer capsules.

In both types, each capsule could be separately swept by gas and its temperature controlled. The three-capsule arrangement was used for the irradiation test. The capsules were numbered from the bottom up as capsules A, B and C.

The three-capsule model was preferred for the following reasons:

- The two fuel elements in capsule B could be placed in the reactor in such a way that they would see the maximum neutron flux in the reactor, while the fuel elements in capsules A and C would be in positions where they would see lower but equal neutron flux. This arrangement made it possible to have two different irradiation regions in the same experiment.
- It was possible to monitor fission gas release under closely controlled conditions.
- Good control over thermo-hydraulic parameters based on previous irradiation measurements HFR-K4.
- Good control over temperatures in the capsules that made temperature cycling possible.

The following requirements had to be met by the irradiation rig during the irradiation experiment:

- Full utilization of the temperature calibration data collected during the HFR-K4 experiment to determine the optimal measurement technique for the irradiation test. Using this data made it possible to determine the surface temperatures of the fuel elements very accurately, and to deduce the centre temperatures from the calibration data.
- Regular and accurate measurement (at least once per day per capsule) of the fission gas release coupled with a temperature measurement. This measurement required the highest measure of accuracy of the whole irradiation testing experiment. As the fission product concentrations that had to be measured were extremely low, the measurement required high accuracy on very low absolute values.
- Accurate determination of neutron flux values seen by the fuel elements, because these flux values were used to determine fuel burn-up and to provide data that were be used to calculate R/B ratios for the fuel elements.

5.3.2.2 Test objectives for HTR-Modul equilibrium core

Table 15 contains the core design values for HTR-Modul, which played a role in specifying Phase 2 irradiation tests.

The nominal values in **Table 15** refer to values obtained for 15 fuel cycles which would result in a burn-up of 8.9% FIMA. After each pass through the reactor core, the burn-up of each fuel element is measured to decide whether it can be allowed to circulate through the reactor core once more. The burn-up measurement has a certain error associated with it. To ma ke allowance for this error, the maximum number of cycles was increased by two. Thus all the maximum values in **Table 15** were deduced from these extra two cycles.

As a result of the specific design of HTR-Modul, the above irradiation test values were characterized by low values for fuel element power and fast neutron fluence. These values were based on a weighted neutron spectrum.

For irradiation testing in HFR Petten, a number of irradiation positions with different neutron spectra were available. It was possible to modify the spectra in these positions by using different filler element materials in irradiation rigs.

The so-called Burn-up Dose Diagram for HTR-Modul as well as for a few irradiation positions in HFR are shown in **Figure 7**. Curve 'M' is the calculated curve for HTR-Modul with the points calculated at the end of each fuel cycle. The other curves represent calculations with different filler elements in the irradiation rig for positions H2 and E1 with fuel elements containing 10% enriched uranium.

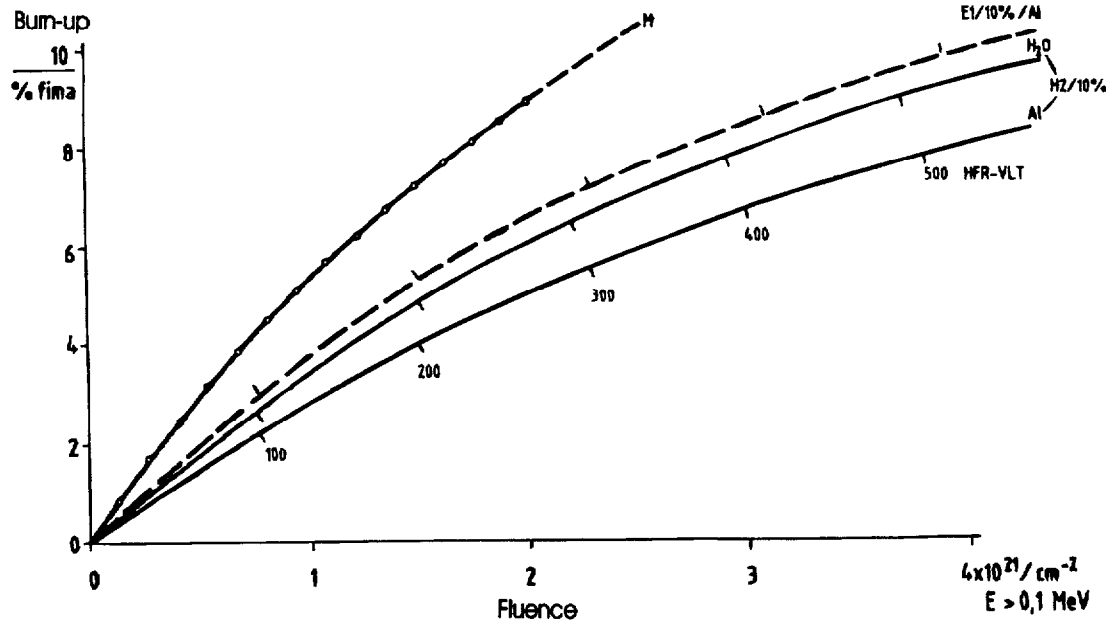


Figure 7: Burn-up vs Fast Neutron Fluence for HTR-Modul

Values for the real enrichment of 8% for HTR-Modul lie between 80% and 85% of the values shown in **Figure 8**.

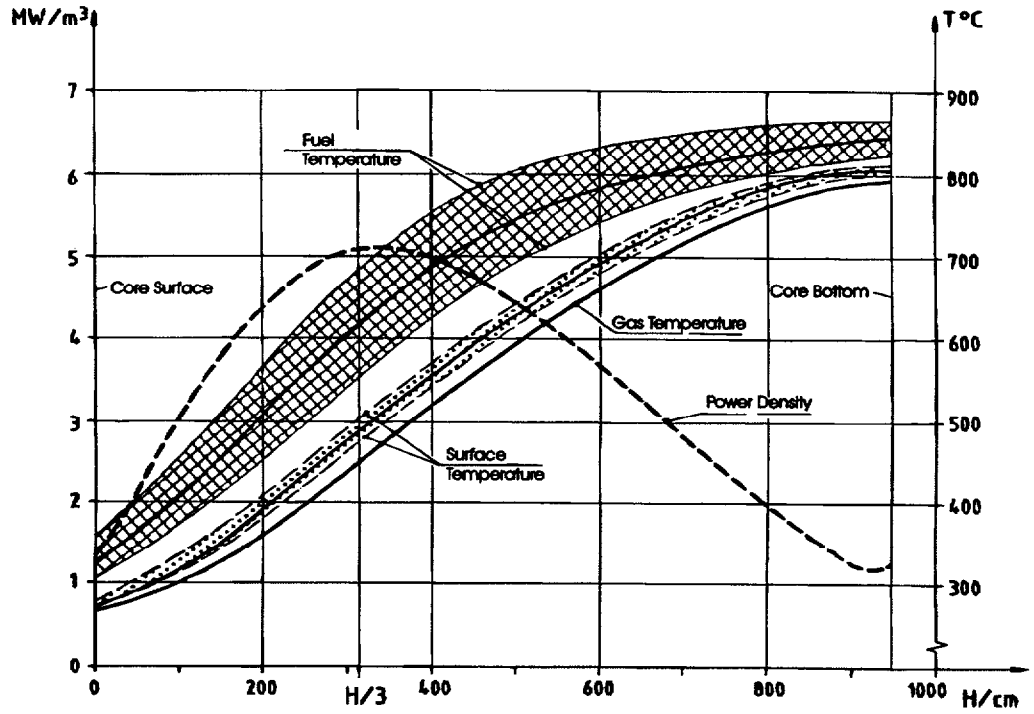


Figure 8: Operating Parameters for HTR-Modul Fuel Elements

Using the curves in **Figure 8**, HFR position E1 with aluminium as the filler element and 10.6% enriched fuel was chosen for the Proof Test. A plan view of the HFR core is shown in **Figure 9** .

A	B	C	D	E	F	G	H	I	
B	B	B	B		B	B	B	B	1
F	F	F		F		F		B	2
F	F		F		F		F	B	3
F	C	F	C	F	C	F		B	4
F	F		F		F		F	B	5
F	C	F	C	F	C	F		B	6
F	F		F		F		F	B	7
F	F	F		F		F		B	8
B	B	B	B		B	B	B	B	9

B	Beryllium Reflector Element
C	Control Assembly
F	Fuel Element
	Irradiation Position

Figure 9: Plan View of HFR Core

Even with this combination, the required fluence values were exceeded by approximately 30%, resulting in a fast neutron fluence of about 3 x 10^{21} cm^{-2}. Experience in preceding irradiation experiments has shown that this excess fast neutron fluence was not critical and could easily be tolerated. Previous irradiation experiments have also shown that enrichment values up to 10.7% could be tolerated by standard TRISO fuel produced in coating batches of 10 kg.

The maximum power per fuel element was 1.4 kW for HTR-Modul, which translated into about 130 mW per coated particle. This value would be exceeded by a factor 2 for the Proof Test in the chosen HFR position. However, in previous irradiation tests with TRISO fuel, the HTR-Modul values have been exceeded by factors between 3 and 4 without any ill eff ects on the fuel integrity.

The chosen irradiation position made it possible to reach the target burn -up within a sufficient time to carry out the planned temperature cycle tests, although the power and fast neutron fluence values expected in HTR-Modul operation would be exceeded.

5.3.2.3 Temperature cycling

With the irradiation test placed in position E1, it was possible to irradiate the test fuel elements for 450 to 500 HFR FPD. Taking into account the availability of the HFR (250 FPD per year in 11 cycles of 23 FPD per cycle), this translated into an irradiation time of approximately two calendar years. Fitting the cycles into HFR cycles could simulate the maximum number of HTR-Modul fuel cycles.

The axial temperature distribution for HTR- Modul is shown in **Figure 8** as a function of reactor height. This temperature could also be related to the time it takes for a fuel element to circulate through the reactor core. The maximum fuel centre temperature of 870 °C is reached at the core bottom on the first cycle. A conservative temperature estimate including all uncertainties of 1 000 °C was used in the Proof Test.

The HFR irradiation cycle was combined with the requirements for HTR-Modul fuel cycle simulation by using a two-phase temperature cycle. A fuel centre temperature of 800 °C was used for one-third of each irradiation cycle (eight FPD) and 1 000 °C for two-thirds of the cycle (FPD) per HFR cycle. This was done for the last 17 HFR cycles. In order to simulate the first HTR-Modul cycle, the test fuel elements were irradiated at a temperature of 800 °C for the first full HFR cycle, and then at 1 000 °C for the following two full HFR cycles.

The difference between the surface temperature of the test fuel element and the expected value in HTR-Modul was small and had no influence on the test parameters.

Simulation of a loss of pressure event was carried out for two test fuel elements during the 1 000 °C part of fuel cycles 5, 11 and 17 (irradiation cycles 7, 13 and 19). During these cycles, the temperature was increased to 1 100 °C for 35 h and then to 1 190 °C for 5 h.

The following arguments support the method of temperature cycling to simulate fuel cycles as described above:

- There is a basic difference between the method of tempe rature cycling and the method of irradiating fuel at a fixed maximum temperature. It has been shown in irradiation experiments performed using HTI-BISO fuel elements that temperature cycling irradiation produced very low particle failure rates, whereas irradiation of the same fuel at constant high temperature produced significantly higher particle failure rates.
-

- The results of test R2-K4 using temperature cycled **HTI**-TRISO particles from an earlier fuel development and test program could not be used as refe rence for the Proof Test. The reason for the massive particle failure rate in that experiment was traced to the relatively high anisotropy of the pyrocarbon coatings for the coated particles used in that test.
- In the HTR-Modul design, the fuel elements are cycled through the reactor core a number of times, and in the process they undergo definite temperature cycles. These temperature cycles give rise to annealing effects and other temperature activated processes in the SiC coating material. This is a major difference between the HTR-Modul design and the design of other HTR reactors such as the HTR-300. Thus the results of previous tests could not be transferred to the HTR-Modul fuel cycle.
- The most important results of the Proof Test would be the results from annealing tests on the irradiated fuel elements. In the heating test, irradiated fuel elements were to be heated to 1 600 °C to simulate a loss of coolant event following a full control rod withdrawal. As a Proof Test for the HTR-Modul, this test would be useless unless the heated fuel elements had been exposed to conditions, including temperature cycling, similar to those expected in the HTR-Modul reactor.

Even if indications were that temperature cycling produced no detrimental effects on TRISO coated fuel, it was still essential to carry out the irradiation tests. The tests would provide experimental proof of this fact, as it is impossible to fully take into account the complex processes taking place during irradiation and temperature cycling in theoretical studies.

5.3.3 Post-irradiation Evaluation

The PIE to be performed was divided into three main groups:

a. General Investigation:
   - Visual Inspection
   - Mass control
   - Dimension measurement
   - Burn-up measurement

b. Fission Product Distribution
   - In test fuel elements
   - In graphite capsule parts
   - In metallic capsule parts for the most important fission products Cs 137/134, Sr 89/90, Ag 110m, I 131

c. Fuel element quality during normal operation and off-normal events
   - Chlorine-leach investigation
   - High temperature annealing tests
   - Ceramographic investigations

As a result of the German HTR programme being stopped while the Proof Test irradiation programme was still in progress, the Proof Test irradiation experiments were completed, but no PIE or heating tests were ever conducted on the irradiated fuel elements.

5.4 Test Results

The test results for HFR-K6 and HFR-K5 are discussed in detail in **[4]**, but will be summarized here. The irradiation data are given in **Table 17** and **Table 18**.

Table 17: HFR-K6 Test Results

		1	2	3	4
Irradiation Time	days	634			
HFR Irradiation Cycles		26			
HTR-Modul Core Passages		17			
Maximum Temperature in Fuel Element Centre					
Steady State	°C	1 090	1 130	1 140	1 130
Transient	°C	1 200	1 250	1 260	1 250
Maximum Power per Fuel Element	kW	1.82	2.51	2.70	2.48
Burn-up	%FIMA	7.2	9.3	9.7	9.2
Fast Neutron Fluence (E > 0.1 MeV)	10^{21} cm^{-2}	3.2	4.6	4.8	4.5
R/B (Kr^{85m} EOL at 1 000 °C)		1.9×10^{-7}	2.1×10^{-7}	2.1×10^{-7}	5.6×10^{-7}

Table 18: HFR-K5 Test Results

		1	2	3	4
Irradiation Time	Days	564.28			
HFR Irradiation Cycles		24			
HTR-Modul Core Passages		17			
Maximum Temperature in Fuel Element Centre					
Steady State	°C	923	909	903	921
Transient	°C	1 013	1 015	1 004	1 001
Maximum Power per Fuel Element	kW	1.32	2.32	2.45	2.36
Burn-up	%FIMA	6.7	8.8	9.1	8.7
Fast Neutron Fluence (E > 0.1 MeV)	10^{21} cm^{-2}	2.85	4.02	4.25	3.91
R/B (Kr^{85m} EOL at 1 000 °C)		1.7×10^{-7}	2.9×10^{-7}	2.9×10^{-7}	3.4×10^{-7}

The target burn-up of 10% FIMA was nearly reached for one of the fuel elements, while the target value for the fast neutron fluence was exceeded by almost a factor two. Target temperature values were achieved during the first six HFR cycles and exceeded by up to 140 °C until the 17th cycle. From the 18th cycle, temperatures were below the target values. Steady fractional release rates for three of the irradiated fuel elements were in the region of 10^{-10} to 10^{-9} initially, and increased by about two orders of magnitude, where it remained for the duration of the measurement. One of the elements showed a different behaviour, increasing to about 10^{-7} to 10^{-6} where it remained steady. From this it was deduced that this fuel element contained two defective particles.

6. OTHER IRRADIATION EXPERIMENTS

Late during the German fuel development programme, three additional fuel elements of the type AVR 21-1 were irradiated in the DIDO reactor in Jülich. This irradiation was designated FRJ2-K15. Due to the termination of the German HTR programme, no PIE was done on these irradiated fuel elements, except for measuring the burn -up values achieved during the test. The test results are shown in **Table 19**.

Table 19: FRJ2-K15 Irradiation Parameters

Parameter	Capsule 1	Capsule 2	Capsule 3
Irradiation Time (EFPD)	651	651	651
Maximum Temperature (°C)	970	1 150	990
Burn-up (% FIMA)	14.1	15.3	14.7
Fast Fluence (x 10^{21} cm^{-2})	0.2	0.2	0.1
Kr-85m R/B Value (EOL)	1 x 10^{-8}	5 x 10^{-9}	3 x 10^{-9}

7. COMPARISON BETWEEN PBMR AND HTR-MODUL

7.1 Introduction

The fuel that will be manufactured for the South African Pebble Bed Modular Reactor (PBMR) is based on the tested and proven German LEU-TRISO fuel design. An extensive and systematic testing programme, described above, backs this fuel design. The German testing programme was aimed at the optimization of the manufacturing process, as well as the testing of the fuel design itself under diverse reactor conditions. These tests were not reactor specific, but generic in nature.

Included in the list of German tests, were experiments to determine particle failure fraction and fission product release from intact and damaged coated particles for a wide range of fuel temperatures and burn-up. The irradiation phase of the generic fuel tests was supplemented by extensive post-irradiation annealing tests to simulate fuel behaviour under conditions expected during reactor transients.

It is not intended to duplicate all the generic work mentioned above in order to qualify locally produced PBMR fuel. The approach will be to duplicate the German manufacturing process and then to show by comparison of physical characteristics that the locally produced fuel is to all intents and purposes equivalent to the German fuel.

Although PBMR fuel will be shown to be equivalent to German fuel, it will have to be qualified in order to prove that it will meet the design requirements for normal operating conditions and anticipated transient conditions specific to PBMR operation. The reactor specific qualification process will proceed in two phases:

- During Phase 1, each step in the manufacturing process will be qualified separately by an extensive programme of measurements to prove that each step in the process delivers an acceptable product. The emphasis during this part of the qualification process will be on proving that the fuel produced conforms to German fuel specifications. This part of the specific fuel qualification programme will be performed by the fuel manufacturing group, and will not be discussed any further in this document.
- During Phase 2 of the qualification process, the emphasis shifts toward proving that the locally manufactured fuel will perform according to PBMR design specifications. Irradiating a number of fuel elements and graphite elements selected randomly from a normal production lot from the manufacturing line will do this. The test fuel elements will probably be taken from the plant qualification production lot.

7.2 Comparison of Specifications

Fuel specifications for German fuel used in Proof Test and for PBMR fuel as contained in PBMR Manufacturing Specifications for Equilibrium Core fuel elements are compared in **Table 20** .

Table 20: Comparison of German Proof Test Fuel and PBMR Fuel Manufacturing Specifications

Property	Unit	Proof Test Fuel Nominal Manufacturing Specification [x] = Average Value	PBMR Manufacturing Specification for Equilibrium Core
Kernel:			
UO_x		≤ 2.015	**Proprietary**
U235 Enrichment	%	10.7 ± 0.1	8 ≤ x ≤ 10 (± 0.10)
Diameter	µm	500	**Proprietary**
Density	g/cm^3	≥ 10.4	**Proprietary**
Coated Particle:			
Buffer Layer:			
Thickness	µm	95	95
Density	g/cm^3	≤ 1.05	≤ 1.05
Inner Pyrocarbon Layer:			
Thickness	µm	40	40
Density	g/cm^3	1.80 ≤ [x] ≤ 2.00	1.80 ≤ [x] ≤ 2.00
Anisotropy (BAF)		≤ 1.10	**Proprietary**
SiC Layer:			
Thickness	µm	35	35
Density	g/cm^3	≥ 3.18	≥ 3.18
Outer Pyrocarbon Layer:			
Thickness	µm	40	40
Density	g/cm^3	1.80 ≤ [x] ≤ 2.00	1.80 ≤ [x] ≤ 2.00
Anisotropy (BAF)		≤ 1.10	**Proprietary**
Fuel Element:			
Matrix Material		A3-3	A3 or equivalent
Thickness of Fuel-free zone	mm	$(\geq 5.0)_{95}$ $(\geq 4.5)_{100}$	$(\geq 5.0)_{95}$ $(\geq 4.5)_{100}$
Thermal Conductivity (1 000 °C)	W/(m.K)	≥ 25	**Proprietary**
Standard Corrosion (1 000 °C)	mg/(cm^2.h)	≤ 1.3	**Proprietary**
Thermal Expansion Anisotropy (20 to 500 °C)		≤ 1.3	**Proprietary**
Crushing Strength	kN	≥ 18	**Proprietary**

Property	Unit	Proof Test Fuel Nominal Manufacturing Specification [x] = Average Value	PBMR Manufacturing Specification for Equilibrium Core
Drop Strength	Number	≥ 50	**Proprietary**
Abrasion	mg/h	≤ 6	**Proprietary**
Heavy Metal Content	g/FE	9.45	9.0
U235 Enrichment	%	10.7	$8 \leq x \leq 10$
Failed Particle Fraction (Burn-leach)		$\leq 6 \times 10^{-5}$	$\leq 6 \times 10^{-5}$
Impurities (Boron Equivalent)	ppm B	≤ 1.3	**Proprietary**

The specifications identified for PBMR fuel are almost identical to those of German reference fuel. These properties are determined by requirements for each reactor design and will be determined during the detail core design phase. As in the case of German Phase 2 tests, it might be necessary to use an enrichment and/or heavy metal loading different from that for the equilibrium PBMR core for irradiation qualification tests. This will be determined by requirements imposed by the materials testing reactor that will be used for fuel irradiation qualification testing.

7.3 Comparison of Normal Operating Conditions

Irradiation load during normal operation can be specified in terms of three parameters: temperature, fast neutron fluence and burn-up. It must be kept in mind that irradiation loads on fuel elements are dependent on core design and will change as the design evolves. The values in this report are specific to the 268 MW core design.

7.3.1 Temperature

7.3.1.1 Graphite element temperature

A basic difference between HTR-Modul and PBMR is that the PBMR reactor core has a central column consisting of graphite spheres that the HTR-Modul reactor core did not have. There is also a so-called mixing zone between the central column and the fuel zone in the PBMR reactor core where mixing of graphite elements and fuel elements occurs. Thus irradiation loads on graphite spheres must be considered for PBMR operations.

The maximum centre temperature for graphite spheres is reached in the mixing zone. **Figure 10** shows the variation of graphite sphere centre temperature along the core height in the mixing zone where the maximum temperature occurs **[5]**.

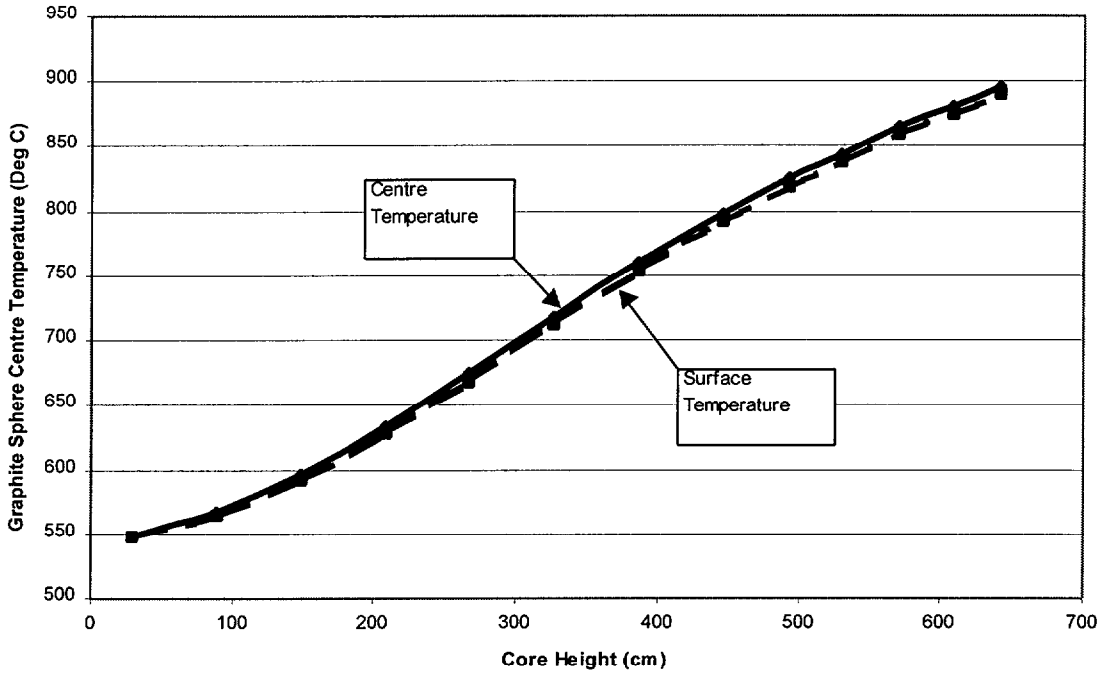


Figure 10: Maximum Centre and Surface Temperatures for PBMR Graphite Spheres

7.3.1.2 Fuel element temperature

Figure 11 [3] and **Figure 12 [5]** show the maximum fuel element centre temperatures for HTR - Modul and for PBMR respectively. For HTR- Modul, the maximum temperature reached during the first fuel cycle was 870 °C. Taking into account all uncertainties, a conservative upper limit of 1 000°C was assumed for maximum fuel centre temperature for HTR- Modul **[3]**.

The nominal maximum fuel element centre temperature for PBMR is 1 054 °C, which, with the same conservatism as used for HTR-Modul, yields a conservative maximum temperature for PBMR of 1 184 °C. **Figure 13** is a plot of the maximum temperature (fuel element centre) as a function of relative core height for HTR- Modul and PBMR.

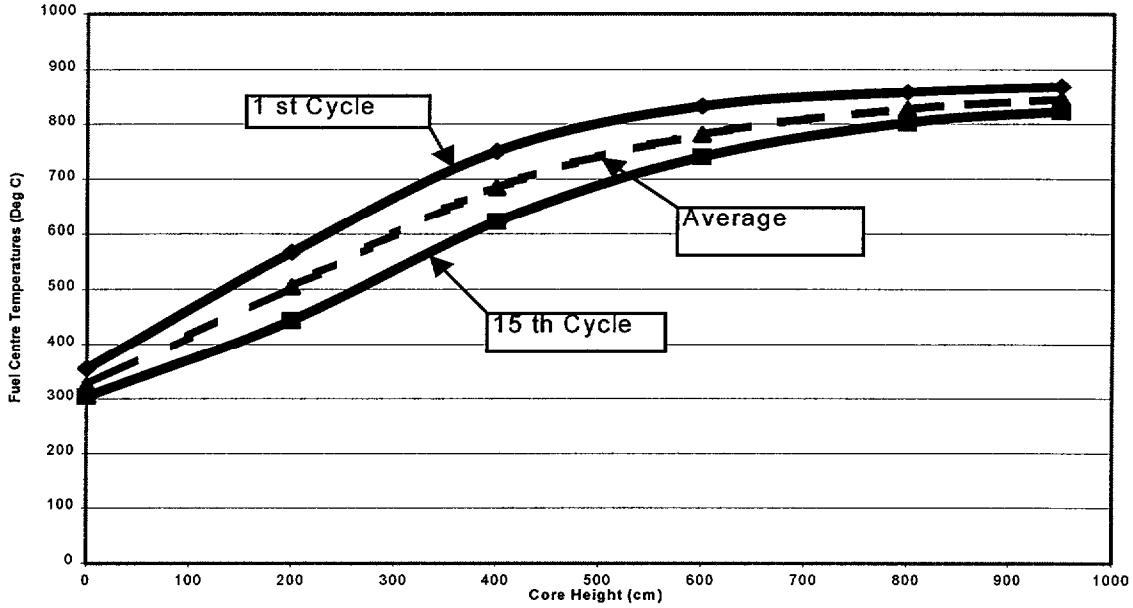


Figure 11: Maximum Fuel Element Centre Temperatures for HTR-Modul

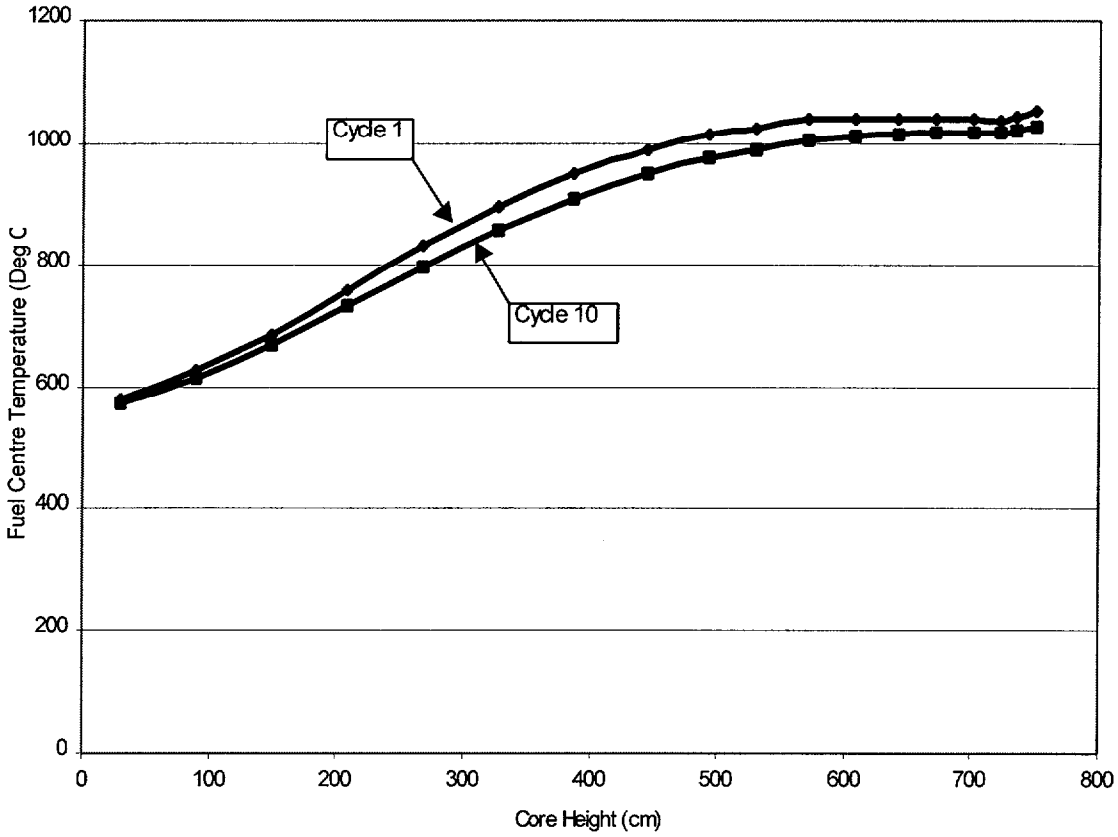


Figure 12: Maximum Fuel Element Centre Temperature for PBMR

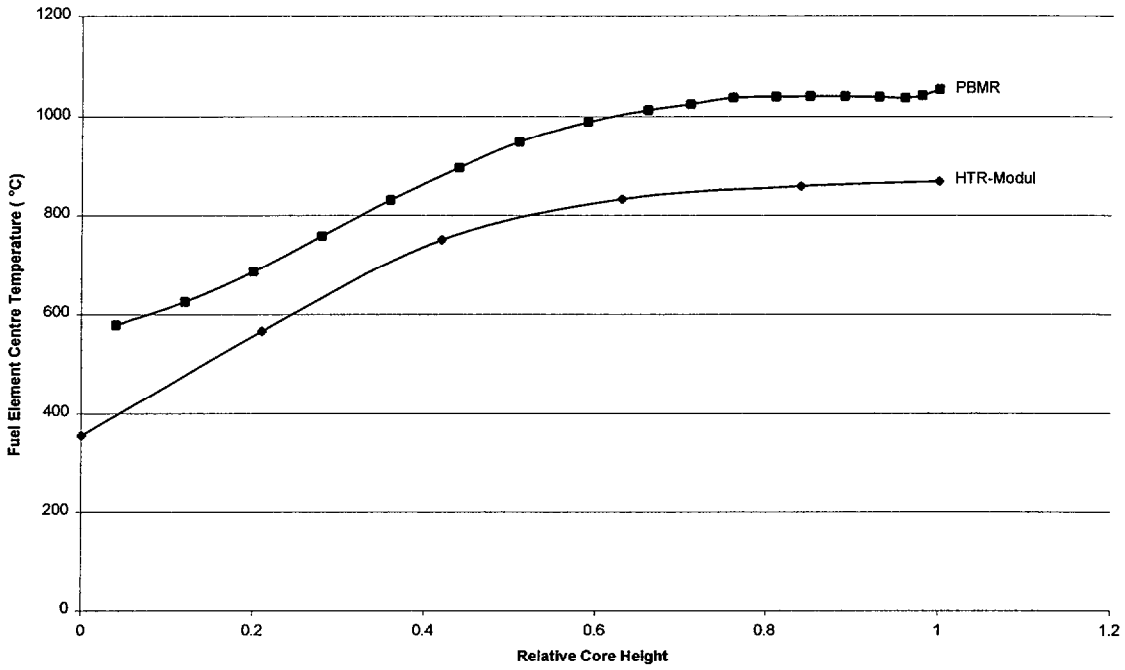


Figure 13: Maximum Fuel Centre Temperatures for HTR- Modul and PBMR (Normalized Core Height)

In HTR-Modul and in the PBMR, fuel elements are inserted at the top of the reactor core, move downwards through the core, are removed at the core bottom and recirculated to the top of the core again. During this process the fuel element passes through regions having different temperatures; low temperature at the top of the core and high temperature at the bottom of the core. In order to simulate HTR-Modul fuel temperature cycles in Phase 2 tests, fuel elements were irradiated using the simple two-step tempe rature cycle shown in **Figure 14**. Fuel elements were kept at 800 °C for one-third of total irradiation time and at 1 000 °C for the remaining two-thirds. The specific characteristics of the materials testing reactor that was used for the experiment determined exactly how this was accomplished.

In the case of HFR Petten, the reactor cycle is 28 days. In order to simulate the first HTR -Modul temperature cycle, fuel elements were irradiated at 800 °C for a whole HFR cycle, and then at 1 000 °C for two full HFR cycles. After that, the pattern was repeated with one-third of an HFR cycle at 800 °C and two-thirds at 1 000 °C.

In order to simulate PBMR fuel cycle temperatures, PBMR test fuel elements will be irradiated at 850 °C for one-third of the irradiation time, and at 1 100 °C for the remaining two- thirds of the time as shown in **Figure 15**. The core lifetime for PBMR fuel elements is approximately 880 FPD and one cycle through the core takes approximately 88 days. The reactor cycle for HFR is 28 days and for IVV-2M it is 21 days.

To simulate the first PBMR fuel cycle will require approximately 29.3 days at 850 °C and 58.7 days at 1 100 °C. This translates into one cycle at 850 °C for both HFR and IVV -2M. Irradiation at 1 100 °C will require two cycles in HFR and three cycles in IVV-2M. Thereafter the temperature will be kept at 850 °C for one -third of each test reactor cycle and at 1 100 °C for the other two-thirds of each cycle.

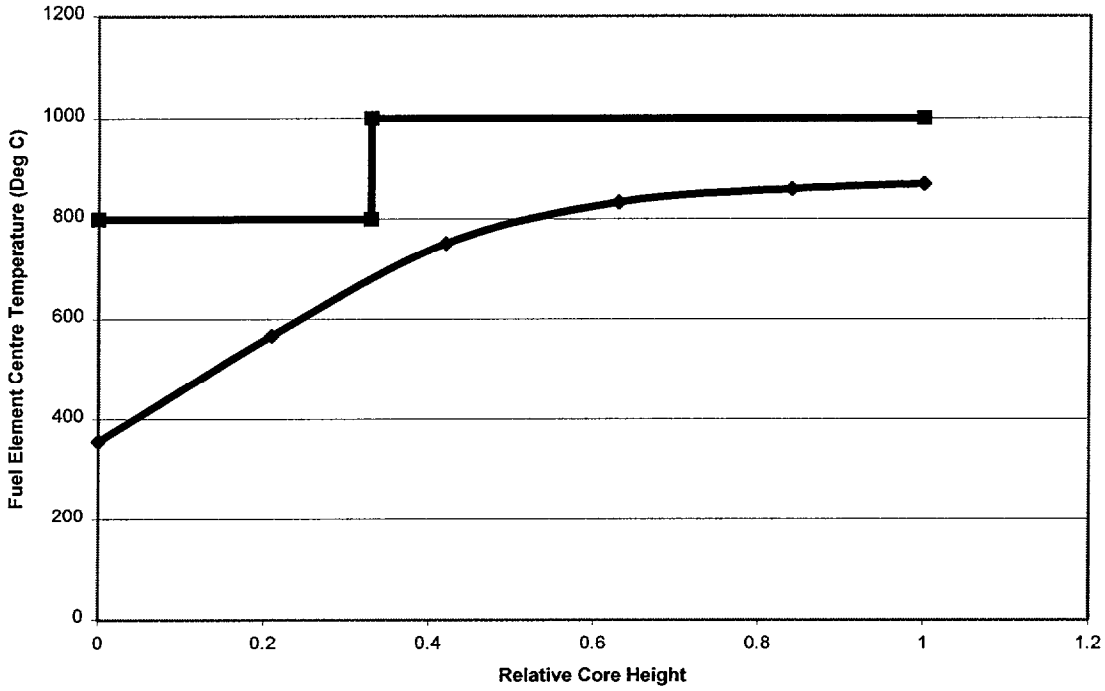


Figure 14: Fuel Element Temperature Cycle for HTR-Modul

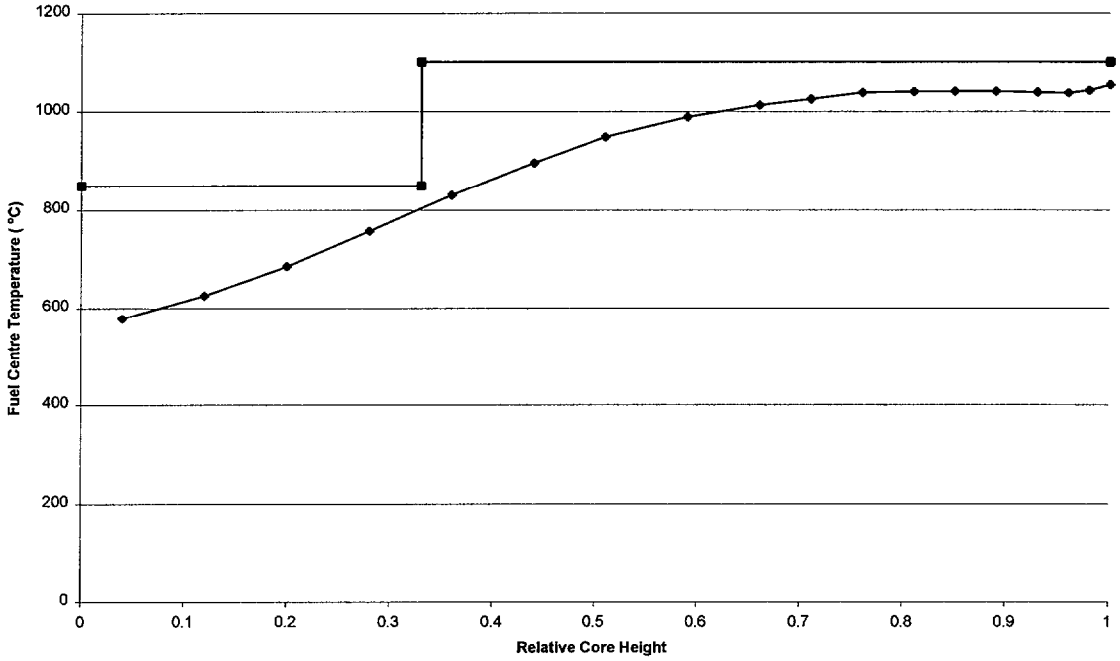


Figure 15: Fuel Temperature Cycles for PBMR

7.3.2 Fast neutron fluence and burn-up

Due to the fact that these two quantities are interrelated to a certain ex tent, they are considered together when defining an irradiation load for fuel irradiation testing. The planned and realized (HFR-K6) irradiation profiles for HTR-Modul **[3]**, **[4]** are shown in **Figure 16**.

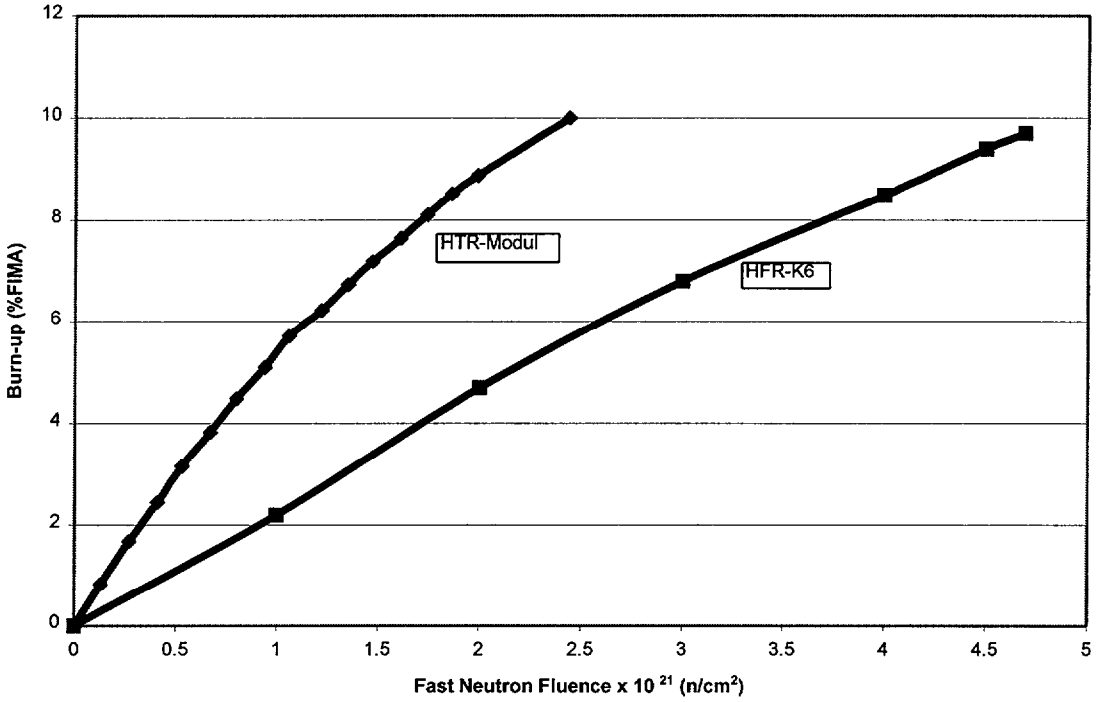


Figure 16: HTR-Modul Target Irradiation Loads

It is clear from the graph that although the target value of 10%FIMA was nearly reached (9.7% FIMA), the target value for fast neutron fluence of 2.4 x 10^{21} n/cm^2 was exceeded by a factor of almost 2 (4.68 x 10^{21} n/cm^2).

The target irradiation load curve for PBMR **[6]** is shown in **Figure 17**. The burn-up target for PBMR is 90 000 MWD/tU (9.36% FIMA) at a fast neutron fluence of 1.68 x 10^{21} n/cm^2 EDN (2.8 x 10^{21} n/cm^2 E>0.1 MeV). Note that this target would be achieved after 12 PBMR fuel cycles. The extra two cycles are introduced to make provision for the expected inaccuracy in burn-up values as measured by the burn-up measurement system.

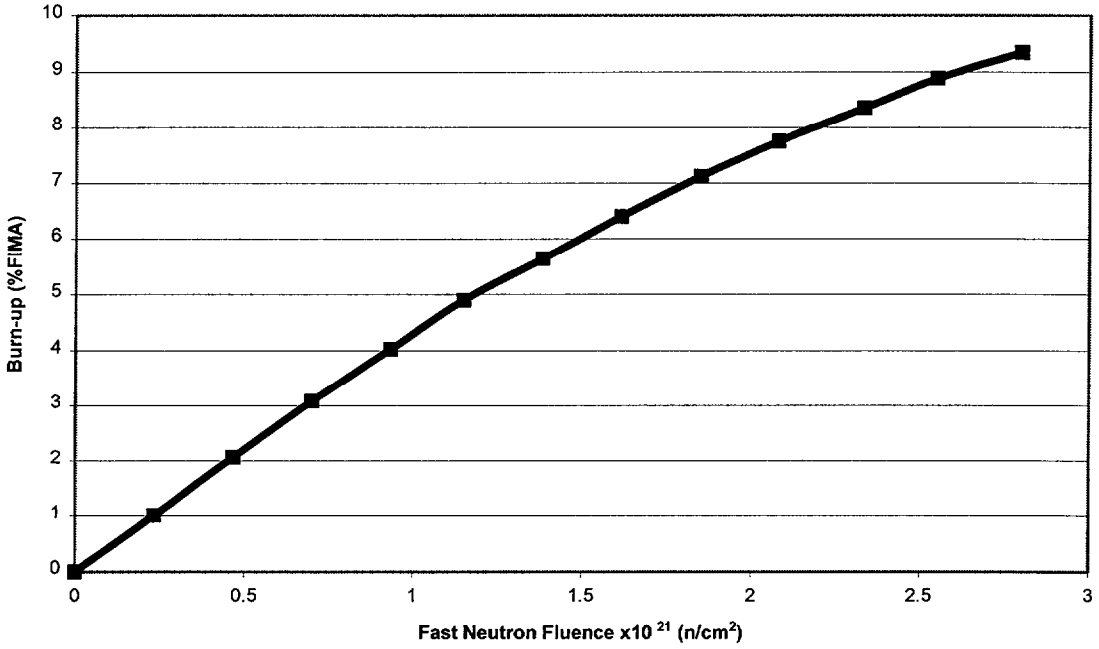


Figure 17: PBMR Fuel Irradiation Load Target

7.4 Comparison of Upset Event Conditions

The upset events tabulated in **Table 21** were considered as design basis events for HTR Modul **[4]**.

Table 21: Maximum Temperatures and Times for HTR-Modul Upset Events

Design Basis Upset Event	Fuel Temperature Increase (°C)	Time to Maximum Temperature (min)	Maximum Fuel Temperature (°C)
Withdrawal of all control rods at full power	100	1	980
Withdrawal of all control rods at 50% power	300	3	1 070
Long-time loss of auxiliary power and emergency diesel unavailability	250	360	1 130
Depressurization with core heat-up	640	1 800	1 520

Maximum temperatures and times for PBMR fuel elements, according to design calculations, are shown in **Table 22** for two important upset events. The two events are Pressurized Loss of Forced Cooling (PLOFC) and Depressurized Loss of Forced Cooling (DLOFC).

Table 22: Maximum Fuel Element Centre Temperatures and Times for PBMR Upset Events

Upset Event	Fuel Temperature Increase (°C)	Time to Maximum Temperature (min)	Maximum Fuel Temperature (°C)
PLOFC	185	510	1 200
DLOFC	519	2 880	1 534

There is some difference between the times to reach the end of the temperature ramp in the PLOFC event for HTR-Modul and PBMR. In the case of HTR- Modul, the maximum temperature of 1 130 °C was reached in 6 h, while the maximum temperature of 1 200 °C for the PBMR is reached after 8.5 h. The slower rise in temperature for PBMR is due to the fact that the central graphite column, which is initially at a lower temperature than the fuel region, acts as a temporary heat sink, restricting the initial rise in fuel temperature.

The conditions of afterheat removal via surface coolers in the case of failure of the main heat sink under pressurized conditions (1 130 °C case) was taken as the basis for upset event simulation for HTR-Modul during irradiation **[4]**. It can be seen from **Table 21** that both control rod withdrawal scenarios were covered by this choice. Thus fuel element centre temperatures were raised from the normal irradiation test maximum of 1 000 °C to 1 200 °C for periods of 3 to 5 h at a time. These transients were introduced at the beginning, middle and end of lifetime for each fuel element during irradiation.

Raising the irradiation temperature from the normal maximum of 1 100 °C to 1 300 °C for 10 h will simulate the PLOFC transient for the PBMR. Transients will be introduced at the beginning, middle and end of lifetime for each fuel element during irradiation.

In the German fuel-testing programme, the DLOFC event was never simulated during irradiation of fuel elements in materials testing reactors. The simulation of this transient was always performed in a separate facility, KÜFA or A-test apparatus, after irradiated fuel elements had been removed from irradiation rigs. This was done to facilitate the measurement of the release of fission products of radiological importance, which could not be done in-pile. For HTR- Modul, it was planned to anneal the irradiated fuel elements at a temperature of 1 620 °C, taking into account the uncertainties by adding 100 °C to the calculated maximum temperature of 1 520 °C.

The temperature history for the core heat-up event (DLOFC) for HTR-Modul is shown in **Figure 18**, together with the temperature history of the so-called standard annealing test procedure that was used to simulate the event in the KÜFA facility **[7]**. The standard annealing test procedure consisted of the following steps:

a. Heating from room temperature to 300 °C in 0.5 h and holding that temperature for typically 5 h to clean the helium circuit and to remove moisture from graphite components.
b. Heating from 300 °C to 1 050 °C in 1.5 h and holding that temperature for typically 15 h.
c. Heating from 1 050 °C to 1 250 °C in 0.5 h and holding that temperature for typically 23 h.
d. Heating from 1 250 °C to the final annealing temperature at a rate of 47 °C per hour and holding the final annealing temperature for up to 500 h.

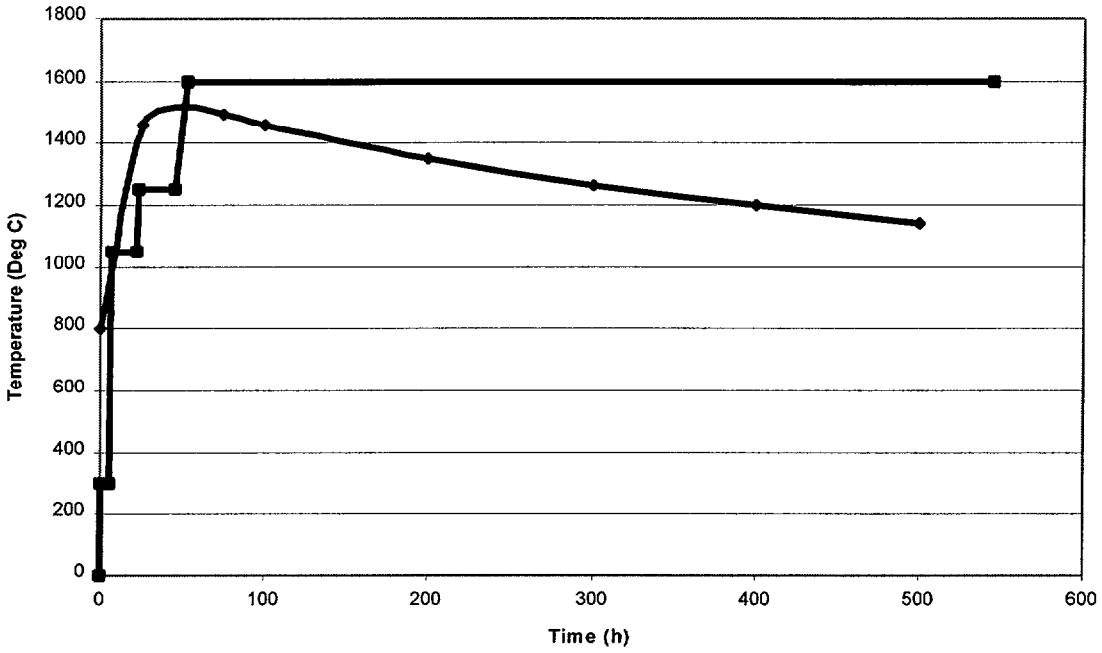


Figure 18: Temperature History for HTR-Modul DLOFC Event and Standard Annealing Procedure

The purpose of the holding points at 1 050 °C and 1 250 °C was to allow fuel and fission products to reach conditions close to those encountered under normal operating conditions in a reactor. During the times spent at the holding temperatures, fission product release fractions at conditions close to normal operating conditions were measured.

The temperature histories for a DLOFC event are compared in **Figure 19** for HTR-Modul and PBMR. The curves are very similar, except for the faster initial temperature rise for HTR- Modul, which could be explained in terms of the presence of the central graphite column in PBMR as above. Thus the standard annealing test procedure described above will be followed for ex -pile annealing of irradiated PBMR fuel elements during fuel qualification. The maximum annealing temperature would be 1 630 °C instead of 1 620 °C as intended for HTR- Modul.

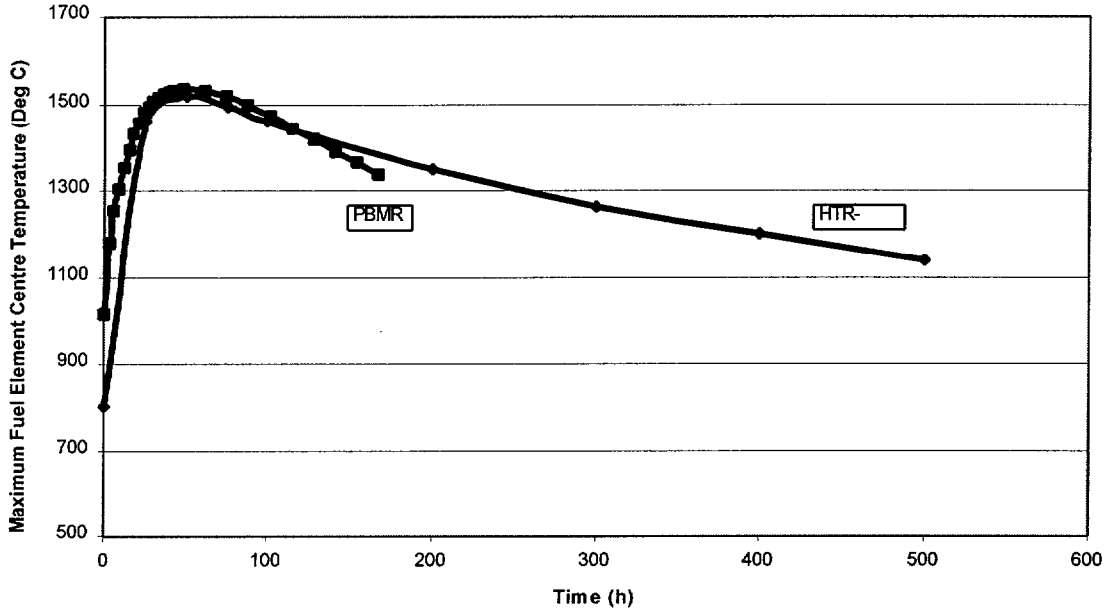


Figure 19: Comparison of Maximum Fuel Element Centre Temperatures Following a DLOFC Event

Table 23 contains a summary of HTR-Modul and PBMR design and test parameters as determined in preceding paragraphs of this report.

Table 23: Summary of HTR-Modul and PBMR Design and Test Values

Property	Unit	HTR-Modul Design	HTR-Modul Test	PBMR Design	PBMR Test
Maximum Burn-up	% FIMA	9.8	10	8.52	9.36
Maximum Fast Neutron Fluence	x 10^{21} n/cm^2	2.4	2.4	2.33	2.8
Maximum Fuel Element Centre Temperature:					
Normal Operation	°C	926	800/1 000	1 054	850/1 100
PLOFC	°C	1 130	1 200 (3 to 5 h)	1 200	1 300 (10 h)
DLOFC	°C	1 520	1 620	1 534	1 634
Maximum Graphite Sphere Centre Temperature	°C	-	-	890	850/1 100
Maximum Power per Fuel Element	kW	1.6	$2 \leq P \leq 3.6$	2.19	$2 \leq P \leq 3.6$
Maximum Power per Coated Particle	mW	138	$130 \leq p \leq 250$	146	$130 \leq p \leq 250$
Number of Core Passes	Number	15	17	10	12

Although PBMR temperatures are generally higher than HTR-Modul temperatures, there is not a single instance where PBMR test parameters and thus normal operation parameters, will threaten the integrity of the fuel elements.

8. APPLICABILITY OF LEU-TRISO TEST RESULTS TO PBMR

In the preceding chapter, PBMR parameters under normal operation conditions and under upset event conditions were compared with HTR-Modul parameters to establish applicable test parameters for PBMR. In this chapter, Phase 1 test conditions will be investigated to establish to what extent the test results are applicable to PBMR fuel elements.

8.1 Temperatures

Table 24 [4], [8] contains temperature, burn-up and fast neutron fluence values obtained in Phase 1 tests on LEU-TRISO fuel elements, AVR, and also the expected values for PBMR. Details of annealing tests performed on these fuel elements are also included. The results of PIE and heating tests performed on some of these fuel elements are described in detail in **[7]**.

Table 24: Test Parameters for Phase 1 Tests and PBMR

Test Number	Sample Number	Irradiation Temperature (°C)	Burn-up (%FIMA)	Fast Neutron Fluence (10^{21} n/cm^2)	Annealing Temperature (°C)	Annealing Time (h)
HFR-P4	1/1	940	13.2	7.0	-	-
	1/2	940	14.1	7.3	-	-
	1/3	940	14.0	7.6	-	-
	1/4	940	14.3	7.8	-	-
	1/5	940	14.3	8.0	-	-
	1/6	940	14.7	7.9	-	-
	1/7	940	14.4	7.5	-	-
	1/8	940	13.8	7.2	1 600	304
	1/9	940	13.8	7.0	-	-
	1/10	940	11.6	6.5	-	-
	1/11	940	11.9	6.1	-	-
	1/12	940	11.1	5.5	1 600	304
	2/1	945	13.6	7.0	-	-
	2/2	945	14.0	7.3	-	-
	2/3	945	14.0	7.6	-	-
	2/4	945	14.0	7.8	-	-
	2/5	945	14.1	8.0	-	-
	2/6	945	14.9	7.9	-	-
	2/7	945	14.1	7.5	-	-
	2/8	945	13.8	7.2	1 600	304
	2/9	945	12.6	7.0	-	-
	2/10	945	12.0	6.5	-	-
	2/11	945	11.3	6.1	-	-
	2/12	945	9.6	5.5	-	-
	3/1	1 075	12.5	7.0	-	-
	3/2	1 075	12.9	7.3	-	-

Test Number	Sample Number	Irradiation Temperature (°C)	Burn-up (%FIMA)	Fast Neutron Fluence (10^{21} n/cm^2)	Annealing Temperature (°C)	Annealing Time (h)
	3/3	1 075	13.3	7.6	-	-
	3/4	1 075	14.7	7.8	-	-
	3/5	1 075	14.0	8.0	-	-
	3/6	1 075	14.0	7.9	-	-
	3/7	1 075	13.9	7.5	1 600	304
	3/8	1 075	13.1	7.2	-	-
	3/9	1 075	12.6	7.0	-	-
	3/10	1 075	12.5	6.5	-	-
	3/11	1 075	10.9	6.1	-	-
	3/12	1 075	9.9	5.5	1 800	279
SL-P1	1	743	8.63	5.0	-	-
	2	750	9.19	5.4	-	-
	3	759	10.01	5.8	-	-
	4	785	10.63	6.2	-	-
	5	788	10.88	6.5	-	-
	6	790	10.69	6.7	1 600	304
	7	793	11.26	6.8	-	-
	8	794	11.07	6.6	-	-
	9	794	10.69	6.3	1 700	304
	10	794	10.32	6.0	1 700	304
	11	780	10.38	5.7	-	-
	12	763	9.51	5.2	-	-
HFR-K3	1	1 200	7.5	4.0	1 600	500
	2	920	10.0	5.8		
	3	920	10.6	5.9	1 800	100
	4	1 220	9.0	4.9		
FRJ2-K13	1	1 125	7.5	0.2		
	2	1 150	8.0	0.2	1 600	138
	3	1 150	7.9	0.2		
	4	1 120	7.6	0.2	1 600	138
AVR 71/22		Surface temp. 1 000->1 280 °C	3.5	0.9	1 600	500
AVR 82/20		Surface temp. 1 000->1 280 °C	8.6	2.4	1 600	100
AVR 82/9		Surface temp. 1 000->1 280 °C	8.9	2.5	1 600	500
AVR 89/13		Surface temp. 1 000->1 280 °C	9.1	2.6	1 620 DLOFC simulation	~10
AVR 85/18		Surface temp. 1 000->1 280 °C	9.2	2.6	1 620 DLOFC simulation	~10

Test Number	Sample Number	Irradiation Temperature (°C)	Burn-up (%FIMA)	Fast Neutron Fluence (10^{21} n/cm^2)	Annealing Temperature (°C)	Annealing Time (h)
AVR 90/5		Surface temp. 1 000->1 280 °C	9.2	2.7	1 620 DLOFC simulation	~10
AVR 90/2		Surface temp. 1 000->1 280 °C	9.3	2.7	1 620 DLOFC simulation	~10
AVR 90/20		Surface temp. 1 000->1 280 °C	9.8	2.9	1 620 DLOFC simulation	~10
AVR 91/31		Surface temp. 1 000->1 280 °C	9.0	2.6	1 700	~10
AVR 74/11		Surface temp. 1 000->1 280 °C	6.2	1.6	1 700	184.5
AVR 88/33		Surface temp. 1 000->1 280 °C	8.5	2.3	1 600/1 800	50/20
AVR 88/15		Surface temp. 1 000->1 280 °C	8.7	2.4	1 600/1 800	50/20
AVR 76/18		Surface temp. 1 000->1 280 °C	7.1	1.9	1 800	200
AVR 88/41		Surface temp. 1 000->1 280 °C	7.6	2.0	1 800	24
AVR 73/12		Surface temp. 1 000->1 280 °C	3.1	0.8	1 900	100
AVR 71/7		Surface temp. 1 000->1 280 °C	1.8	0.5	2 000	100
AVR 70/19		Surface temp. 1 000->1 280 °C	2.2	0.6	2 400	Ramp
AVR 74/8		Surface temp. 1 000->1 280 °C	2.9	0.7	2 500	Ramp
AVR 76/19		Surface temp. 1 000->1 280 °C	7.3	1.9	1 900	30
AVR 80/22		Surface temp. 1 000->1 280 °C	9.1	2.4	1 900	30
AVR 80/16		Surface temp. 1 000->1 280 °C	7.8	2.0	2 000	30
AVR 74/6		Surface temp. 1 000->1 280 °C	5.6	1.4	2 100	30
AVR 76/28		Surface temp. 1 000->1 280 °C	6.9	1.8	2 100	30
AVR 76/27		Surface temp. 1 000->1 280 °C	7.4	1.9	2 100	30
AVR 80/14		Surface temp. 1 000->1 280 °C	8.4	2.2	2 500	Ramp
PBMR		980	9.36	2.33	1 630	200

The irradiation temperature distribution of Phase 1 and AVR irradiation tests is shown in **Figure 20**. Of the 81 fuel elements and small spheres tested in Phase 1 and AVR irradiation tests, 43 (53%) were irradiated at temperatures higher than the maximum expected temperature (1 054 °C) in the PBMR. Of the fuel elements irradiated at lower temperatures, 27 (33%) were irradiated at temperatures only 130 °C lower than 1 054 °C. Thus it is clear that PBMR temperatures fit well into the temperature envelope of Phase 1 and AVR irradiation temperatures.

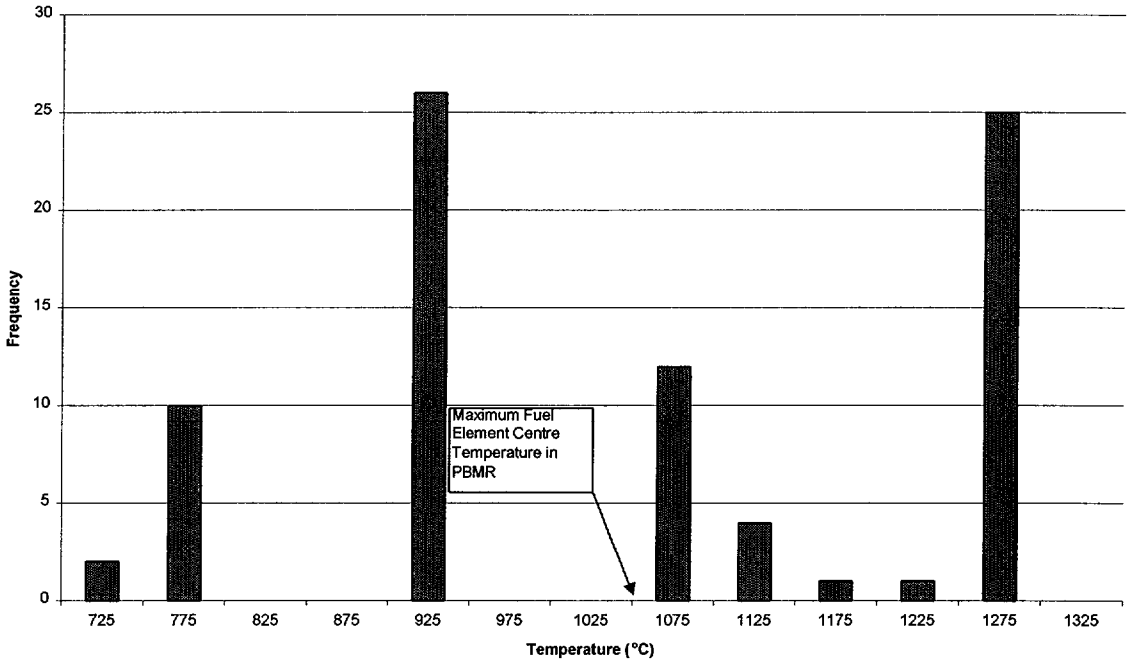


Figure 20: Irradiation Temperature Distribution for Phase 1 and AVR Irradiation Tests on LEU-TRISO Fuel Elements

8.2 Burn-up and Fast Neutron Fluence

Figure 21 shows the irradiation load graphs for PBMR and HTR- Modul together with point values for the irradiation loads for Phase 1 and AVR irradiation tests on fuel elements and small spheres. The point loads occur in two main groups. The first group forms a band just below the HTR-Modul irradiation load curve, while the other group lies at fast neutron fluence values beyond approximately 5 x 10^{21}n/cm^2. It is clear from the graph that expected PBMR irradiation loads fit well within the envelope provided by Phase 1 and AVR irradiation results.

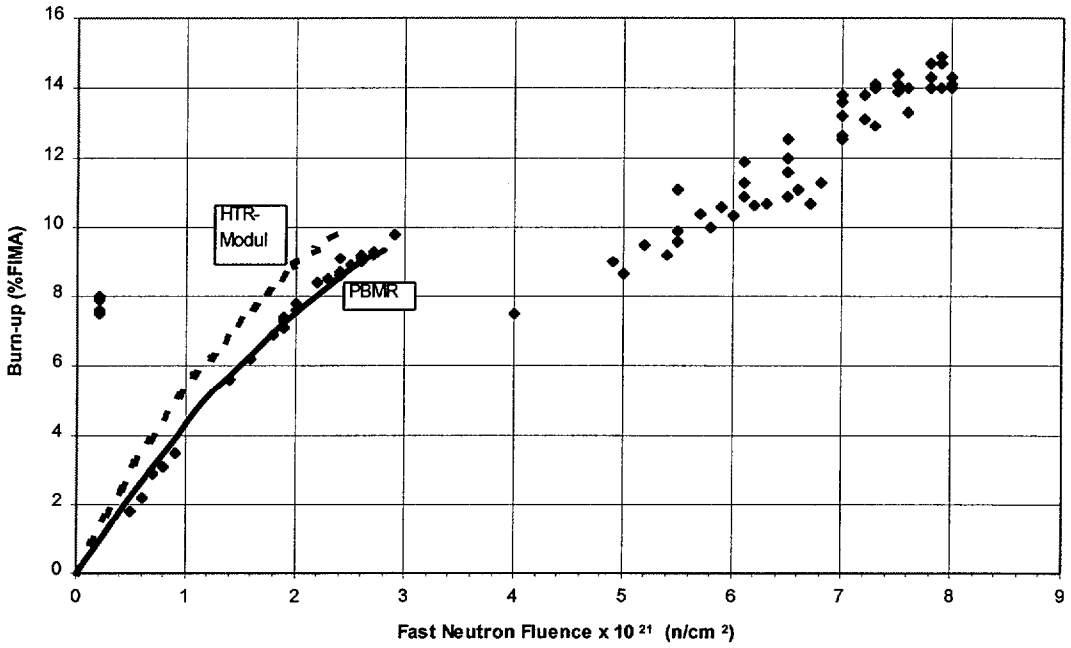


Figure 21: Irradiation Loads for PBMR, HTR-Modul, Phase 1, and AVR Irradiation Tests

It is clear from the above that PBMR normal operation loads (temperature, fast neutron fluence and burn-up) on fuel elements are well represented by Phase 1 and AVR irradiation tests. Thus the data from these tests are applicable to PBMR fuel elements, and it would not be necessary to repeat any generic tests for PBMR fuel, provided that PBMR fuel elements are equivalent to German LEU-TRISO fuel. In defining the fuel ir radiation-testing programme for PBMR, it is assumed that only reactor specific irradiation tests (Proof Tests) need to be done.

9. PBMR FUEL IRRADIATION QUALIFICATION PROGRAMME

This section was intentionally deleted from US NRC version submitted on May 30, 2002.

10. CONCLUSION

This report has described the German qualification programme for LEU-TRISO fuel elements in considerable detail. The result of this programme determines the envelope within which this type of fuel element can be used in a power reactor. It was shown that expected PBMR fuel load requirements fall well within this envelope. Given the PBMR fuel manufacturing strategy of ensuring that PBMR fuel elements will be equivalent to German LEU-TRISO fuel elements, it will not be necessary to repeat the generic enveloping qualification experiments. Thus only a Proof Test will be performed on PBMR fuel and graphite elements to show that PBMR fuel will perform according to requirements in the PBMR core.

PBMR requirements were used to develop a Proof Test Programme for PBMR fuel and graphite elements and to specify test requirements. These test requirements will be used to generate a Proof Test specification.

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